
SKBF TECHNICAL KBS REPORT

83-77

KBS ANNUAL REPORT 1983

**Including Summaries of Technical
Reports Issued during 1983**

KBS Stockholm June 1984

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SUMMARIES OF KBS' TECHNICAL REPORTS 1983
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1 GENERAL BACKGROUND

1.1 THE SWEDISH NUCLEAR POWER PROGRAM

During 1983 10 reactors were in operation generating 37% of the total Swedish electric power production. Two reactors, Oskarshamn 3 and Forsmark 3, were under construction. The Swedish parliament has decided that the nuclear power program is to be limited to 12 reactors and that no nuclear reactor is to be operated after the year 2010. This constitutes the basis for the present nuclear waste management planning in Sweden.

1.2 WASTE TYPES, QUANTITIES AND PRINCIPLES OF HANDLING AND FINAL STORAGE

Following the parliamentary resolution to limit the Swedish nuclear power program, an overall estimate can be made of the quantities of different types of radioactive waste that will have to be disposed of in Sweden.

The maximum quantity of spent fuel is estimated to about 7 500 tonnes (metric tons). Approximately 10% of this amount, or about 730 tonnes, is covered by foreign reprocessing contracts stipulating that most of the radioactive waste from reprocessing is to be returned to Sweden for final storage. At the present time, no further reprocessing of Swedish spent nuclear fuel can be foreseen. In the first place, there is a shortage of available international capacity, and in the second place, it is unclear as to whether reprocessing of spent fuel from the limited Swedish nuclear power program is desirable.

The types and total quantities of various waste categories currently of relevance in Sweden are given in Table 1-1. In addition to these quantities about 5 000 m³ of radioactive wastes

Table 1-1. Waste categories

Waste category	Origin	Waste form	Properties	Quantity
1a Spent fuel	Operation of nuclear reactors	Fuel rods encapsulated in copper canisters	High heat flux and radiation at first. Contains long-lived nuclides	4 840 canisters
1b High-level waste	Residual products from reprocessing	Vitrified waste encapsulated in lead-titanium canisters	High heat flux and radiation at first. Contains long-lived nuclides	730 canisters
2 Transuranic-bearing waste	Waste from the reprocessing process	Solidified in concrete or bitumen	Low- to medium-level. Contains long-lived nuclides	4 500 m ³
3 Core components and internals	Scrap metal inside reactor tanks	Untreated or cast in concrete	Low- to medium-level. Contains certain long-lived nuclides	15 000 m ³
4 Reactor waste	Operating waste from nuclear power plants etc.	Solidified in concrete or bitumen. Compacted waste	Low- to medium-level. Limited life time	116 000 m ³
5 Dismantling waste	From dismantling of nuclear facilities	Untreated for the most part	Low- to medium-level. Limited life time	114 000 m ³

from other sources (industrial and medical use of radioisotopes) will be finally deposited together with the reactor waste according to a special agreement signed between SKBF and STUDEVIK during 1983.

1.3 LEGISLATION AND NATIONAL AUTHORITIES

Activities aimed at a safe handling and final disposal of radioactive waste from nuclear power generation in Sweden are regulated by several Acts. Basic licences are issued by the Government. The processing of licence applications and the enforcement of regulations are handled by various national authorities.

A new Act on Nuclear Activities was proposed by the Government during 1983 and entered into force Febr 1, 1984. This Act replaces the earlier Nuclear Energy Act and the Stipulation Act. The new Act sets forth the basic regulations on construction, possession or operation of nuclear facilities as well as on the possession or handling of nuclear material or nuclear waste.

A special licence is required before fueling a new reactor. It is granted only if the reactor owner (1) has shown to the satisfaction of the Government that there exists an acceptable method for the handling and final disposal of spent nuclear fuel and (2) presented an acceptable plan for the continued research and development needed for the implementation of the waste management program. Updated versions of this plan shall be submitted every third year to the Government or to the authority designated by the Government for examination and evaluation.

The Radiation Protection Act contains regulations aimed at protection against ionizing radiation.

The Environment Protection Act contains regulations aimed at protection of the environment.

The Building Act §136a sets forth stipulations for the siting of so-called "polluting industrial plants".

The municipality has the right of veto.

The Financing Act stipulates that the primary technical and economic responsibility for the disposal of radioactive waste lies with the reactor owner and that the State shall control the arrangements and guarantee the financing of future activities through funds built up from charges levied on nuclear electricity production.

The following national authorities deal with matters connected to radioactive waste management

- The Swedish Nuclear Power Inspectorate, SKI, supervises and controls the design, construction and operation of nuclear facilities,
- The Swedish Radiation Protection Institute, SSI, supervises and controls that appropriate measures for radiation protection are taken by the utilities,
- The National Swedish Franchise Board for Environment Protection is licensing authority as far as other than radiological impacts on the environment are concerned,
- The National Board for Spent Fuel, NAK, supervises and controls the utilities program for R&D and administers the funds for future costs for the radioactive waste management.

1.4 THE POWER INDUSTRY'S ORGANISATION FOR WASTE MANAGEMENT

The New Act on Nuclear Activities mentioned in the preceding section entering into effect on 1984-02-01 regulates the division of responsibility between the State and the power utilities with respect to the handling and final storage of spent nuclear fuel and radioactive waste stemming from such fuel. The Act also covers the dismantling of the nuclear power plants, but not the management of reactor waste that is continuously produced during the operation of the nuclear power plants.

The Swedish legislation explicitly puts the primary responsibility for the radioactive waste management on the nuclear power utilities. The role of the national authorities is mainly to supervise and control that the activities are sufficient, relevant and in accordance with legal requirements. The utilities (The Swedish State Power Board, Sydkraft, OKG and Forsmarks Kraftgrupp) have delegated the execution of the activities in this field to the jointly owned Swedish Nuclear Fuel Supply Company, SKBF¹⁾.

Departments have been organised within SKBF for various tasks. Research and development activities are conducted within the KBS department (Kärnkraftavfallets Behandling och Slutförvaring = Handling and Final Disposal of Nuclear Power Waste). Other departments within SKBF are responsible for the construction and operation of CLAB and for the transportation system. SKBF's and KBS' places in the organisation scheme are shown in Figure 1-1.

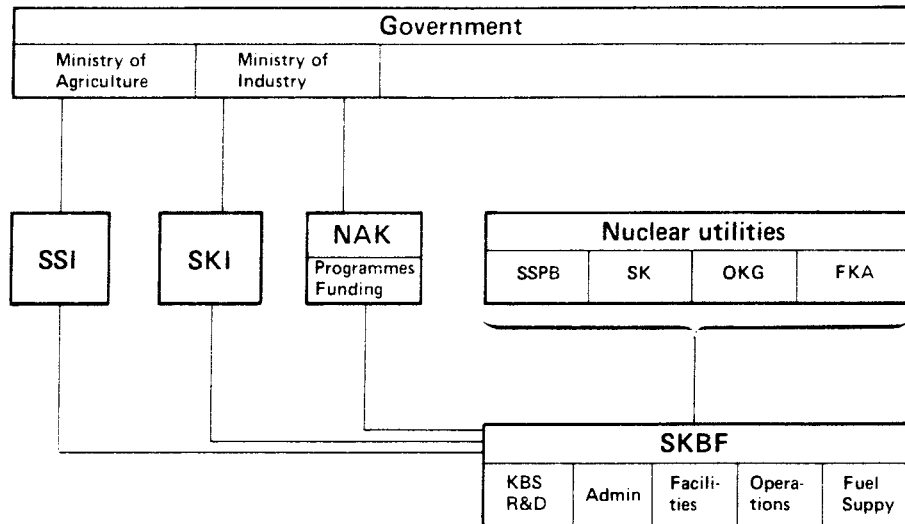
1.5 PROGRAM AND GOAL OF KBS' WORK

1.5.1 General

The scheme in Figure 1-2 illustrates the principle for the handling of assignments within KBS.

KBS was organized in connection with the enactment of the Stipulation Act in 1977 and was assigned the responsibility of conducting such studies and preparing such reports as are required by the Act. The two alternatives offered by the Act - demonstrating how a safe handling and final storage can be effected either for high-level waste from reprocessing or for spent nuclear fuel - have accordingly been dealt with. It has been demonstrated that a safe handling and final storage is possible with present-day

1) From 84-07-01 the name of the company is changed to Swedish Nuclear Fuel and Waste Management Company, SKB.



Controlling authorities:

NAK	National Board for Spent Nuclear Fuel
SKI	Swedish Nuclear Power Inspectorate
SSI	National Institute for Radiation Protection

Utilities:

SSPB	Swedish State Power Board
FKA	Forsmarks Kraftgrupp AB (SSPB 74.5%, private and municipalities 25.5%)
SK	Sydkraft AB (municipalities 53.5%, private 46.5%)
OKG	OKG Aktiebolag (Sydkraft 35%, municipalities 16.25%, private 48.75%)

Executing organization:

SKBF	Swedish Nuclear Fuel Supply Co (SSPB 36%, FKA 30%, OKG 22%, SK 12%)
KBS	A division of SKBF and responsible for R & D

Figure 1-1. Main features of the Swedish organization for the management of radioactive waste.

technology for both alternatives. The description of a reprocessing alternative (1-1) was approved in 1979 by the Swedish Government. A report on the second alternative, direct disposal of spent fuel, (1-2) was submitted to the Government in May 1983 as support for applications to charge the two new reactors, Forsmark 1 and Oskarshamn 3 with fuel. The Government has sent the report for review to 22 Swedish and 8 foreign institutions. Their statements will be available early 1984 and the final decision of the Swedish Government is expected in mid 1984.

The planning, design and start of construction of an underground final repository for reactor waste near the Forsmark nuclear power plant has also been handled within the division KBS during 1983. A construction license was granted by the Government in June 1983 and excavation of the tunnels started in August 1983.

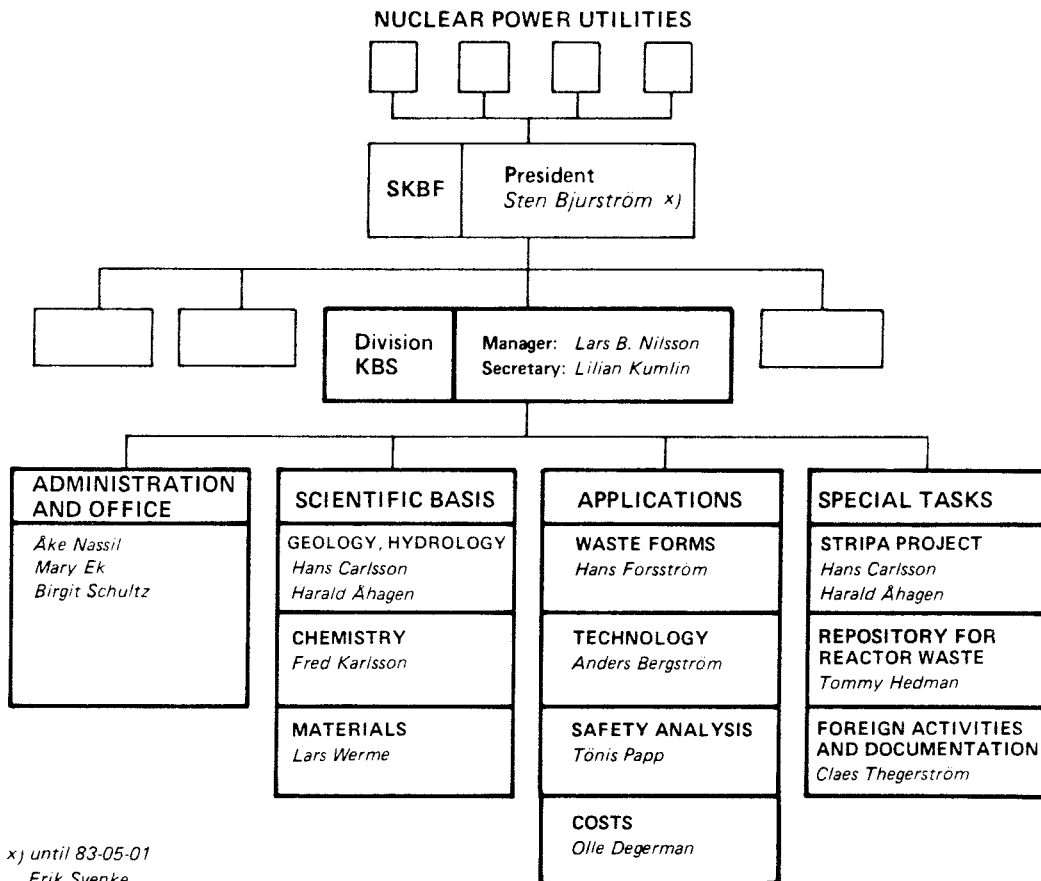


Figure 1-2. KBS organization during 1983.

1.5.2 Long-range program

Even if the KBS reports have shown that a safe final storage of long-lived radioactive waste can be effected with present-day technology and the geological conditions existing in Sweden, considerable work remains to be done to show, how these measures are to be realized in detail and in an optimal way.

The general principles which have been adopted as a basis for the design of a final repository for high-level radioactive waste, and which are deemed to be in compliance with current Swedish legislation in the field, are the following:

- o Siting deep in selected crystalline bedrock in Sweden.
- o The utilization of a multibarrier system, where the barriers consist of the limited solubility of the waste in the groundwater, a canister of a durable material, a layer of "buffer material" between the canister and the bedrock and finally the bedrock itself with low groundwater flow and chemical retardation effects.
- o Application of known technology.
- o No dependency on long-range surveillance for the safety of the repository.

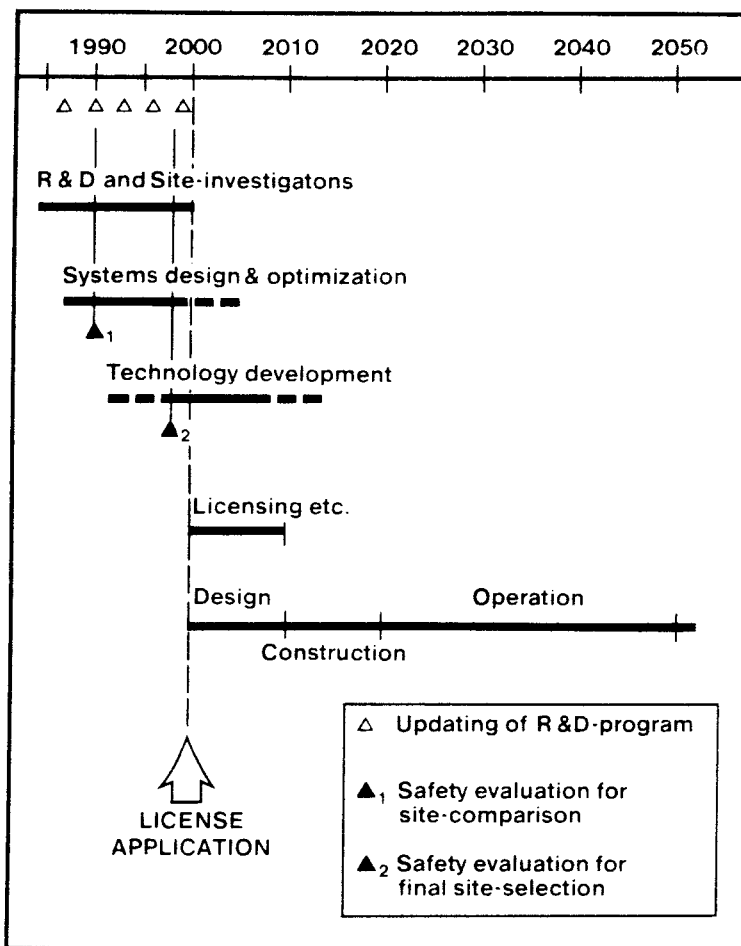


Figure 1-3. General time-table.

The general principles can be assumed to be applicable in the future as well. It is, however, obvious that various solutions within these fundamental frames should, as in the past, continue to be considered and tested in the light of continuous technical-scientific development in Sweden and abroad. Thus, even though definitive designs cannot be given in detail today, it is necessary to base overall efforts and cost calculations on a realistic scenario for the treatment and handling of the waste. The general time-schedule that currently serves as a basis for the work is shown in Figure 1-3.

Owing to the lack of sufficiently detailed data, the safety assessments made up until now have, of necessity, been based in part on pessimistic assumptions. As more and more knowledge is gained, the systems can be defined with greater accuracy. Within the fields of chemistry and hydrology in particular, continued research promises to yield a better understanding of the phenomena that determine the safety of a final repository. Extensive research within these areas is therefore planned for the years to come.

The choice of a site for a final repository for high-level waste can be of technical-economic importance and is also of great political and public interest. Extensive geological investigations

are therefore being planned at many places in the country so that a broad and reliable body of data will be available by the end of the century, see Chapter 6.

1.5.3 Short-range program

During 1983 work started on an updated program for continued R&D. In accordance with the new Act on Nuclear Activities this program is based on the feasibility study presented in KBS-3. During the next few years the program will be revised to cover also other possible designs for the final handling and storage of spent fuel. This revised program will be presented to the Government in the later half of 1986.

During the 1980ies the emphasis will be placed on R&D necessary to provide a basis for the site comparison planned for the turn of the decade.

The Main activities comprise

- Continued site investigations
- Development of a comprehensive computerized base of geodata
- Studies of and models for groundwater flow and radionuclide migration in fissured zones
- Leaching and corrosion of spent fuel
- Natural changes with time of groundwater recipients in the biosphere
- Development of computerized systems for performance assessment.

1.6 INTERNATIONAL COOPERATION

Cooperation and exchange of information with foreign and international organisations has continued during 1983. Among other things, staff members of KBS and experts engaged by KBS have participated in activities within the IAEA and the OECD/NEA.

International development in the field has been followed through participation in a number of conferences, where Swedish papers have also been presented.

The KBS-managed multinational OECD/NEA project at Stripa has progressed according to schedule and a phase 2 of the project has been agreed upon. The following countries are now participating

in the Stripa Project: Canada, Finland, France, Japan, Sweden, Switzerland, United Kingdom and the United States. Spain has declared the intention to join the project.

The original program for the JSS project, a joint investigation on waste glass leaching supported by CRIEPI, Japan, NAGRA, Switzerland and with SKBF/KBS as managing party, is nearly completed. An additional phase to be performed during 1984-85 is under discussion.

Information has been exchanged with US DOE, AECL, Canada and NAGRA, Switzerland according to bilateral agreements. A similar agreement with CEA, France, has been negotiated and is ready for signatures.

KBS has participated in the INTRACOIN-project for comparison of codes for nuclide migration in the geosphere. The intention is also to participate in the planned HYDROCOIN-project concerning comparison of codes for groundwater flow.

2 CHARACTERIZATION AND TREATMENT OF WASTE

2.1 GENERAL

KBS' work on the management and final storage of radioactive waste is primarily concerned with the spent fuel and the waste obtained from the operation of nuclear power plants. The waste types studied are:

Spent fuel reprocessing waste

- high-level vitrified waste
- low- and medium-level waste

reactor waste

decommissioning waste

Besides these types of waste, studies are also being conducted of the possibility to deposit other types of radioactive waste as well - for example waste generated in industry, health care and research - in the final repositories being planned for the nuclear power waste.

Most of the R&D work within KBS has been devoted to the final storage of spent fuel and high-level waste. This work is described in the following chapters. Reactor waste has also been given high priority during the last years with special emphasis on the design of the Final Repository for Reactor Waste (SFR) and the safety assessment for this repository. This is described more in detail in Chapter 8. The work on the final storage of low- and medium-level reprocessing waste and decommissioning waste is still in an introductory phase and is therefore discussed in a brief passage in this chapter.

2.2 WASTE FROM REPROCESSING

2.2.1 General

The contract between SKBF and COGEMA concerning the reprocessing of spent nuclear fuel provides that all waste deriving from reprocessing may be returned to Sweden. In addition to the high-level glass, this also includes several types of low- and medium-level waste. Work on specifying the final form of this waste is in progress within COGEMA, and a first series of specifications for the vitrified waste was submitted in 1982. Specifications for the low- and medium-level wastes are expected in mid 1984. The formulation of these specifications has been discussed at meetings between the COGEMA and its clients. SKBF is being represented by KBS in these discussions.

KBS' work on this waste is primarily aimed at demonstrating that the waste can be managed and finally disposed of in a safe manner. Most of the waste contains long-lived alpha activity. For this reason, the final disposal of the low- and medium-level waste from reprocessing is planned to be coordinated with the final disposal of high-level waste and spent fuel and will therefore not start before 2010. Work on this waste is of a basic R&D character.

2.2.2 Waste types and planned treatment methods

Table 2-1 lists the most important types of waste obtained from reprocessing according to COGEMA and tells how they are planned to be treated at La Hague.

Table 2-1. Waste from reprocessing

Waste type	Treatment method
High-level liquid waste	Vitrification
Fuel cladding hulls and pieces	Solidification with cement
Ion exchange resins and sludge	Solidification with bitumen
Scrap, paper, plastic etc	Solidification with cement

2.2.3 High-level vitrified waste

The vitrified waste that is returned to Sweden will contain nine percent by weight fission products. Most of the work on characte-

rizing this glass is being done in France. At the same time, studies are being conducted in Sweden on how the glass behaves in the final repository environment. This work is discussed in Chapter 3.

2.2.4 Low- and medium-level waste

A question of great importance for the safety assessment of the final disposal of cemented alpha-containing reprocessing waste is the behaviour of actinides at the high pH caused by the concrete. Fundamental experimental studies of this issue are in progress. They comprise of sorption and diffusion measurements for different concrete types. In total seven different concretes have been used, two of which are more than 70 years old. The pore water has been characterized for each sample as regards pH, primary cations, ionic strength, eH etc.

Sorption measurements on crushed concrete in a pore water solution have been performed with Cs, I, Pu, Am(III), Np(V) and U(VI). The results indicate that in general the distribution coefficient for concrete is at least as high as for rock samples at neutral pH. Iodine (I) has a higher (10-100 times) distribution coefficient on concrete than in rock while Cs has a substantially lower distribution coefficient.

The effect of complexing agents such as TBP or triethanolamine was studied in some samples and was found to be low at the concentrations expected. Diffusion measurements in different concrete samples with Cs, Am and Pu are in progress since mid -83 and results are expected during 1984.

The chemical environment can be altered by radiolysis. Theoretical studies of radiolysis in concrete have been performed. The results from simplified calculations have been compared with measurements. It is found that these calculations can reproduce the general behaviour. On certain points, eg. the measured consumption of oxygen during irradiation, however, they fail and a more detailed model is needed.

2.2.5 The HIPOW process

An alternative method for the treatment of cladding hulls, developed by ASEA under contract to KBS, was described in the annual report of 1980. The method is called HIPOW (Hot Isostatic Pressing of Waste) and involves pressing the hulls into a fully compact, pore-free zircaloy block. Pressing takes place in a hot isostatic press at about 1 000°C and 150 MPa. During 1982/83 the studies have been completed with test pressings of non-precompact cladding hulls in a bellow canister.

2.3 REACTOR WASTE

2.3.1 General

Low- and medium-level waste with a hazardous life shorter than 500 years is generated during operation of nuclear power plants and the storage of spent nuclear fuel. This waste is called reactor waste. KBS' work on reactor waste is aimed primarily at developing safe disposal methods. KBS' resources are also utilized for studies of treatment methods for different waste types of common interest to the Swedish nuclear power industry.

Work on a final repository for reactor waste, SFR, has been going on during the year. It is described in Chapter 8. In parallel with this project, studies are also being conducted of the product characteristics of the waste that is to be deposited in the SFR as well as on new treatment methods for the waste.

2.3.2 Waste types and current treatment methods

Table 2-2 lists the most important waste types obtained in connection with reactor operation and tells how they are currently treated.

Table 2-2. Current treatment methods for waste from Swedish nuclear power plants.

Waste type	Current treatment
Core components	No treatment
Ion exchange resins and sludge	Solidification with cement or bitumen Dewatering
Scrap	Incorporation in cement or simply packaging
Low-level solid waste	Compaction or incineration

Currently, this waste is stored at the nuclear power plants pending construction of the central final repository.

2.3.3 Ion exchange resins solidified in cement

Solidification systems for ion exchange resins in cement have been in operation at two Swedish nuclear power plants for about ten years. The operations have generally been successful. The volume increase incurred by the process is, however, disturbing. Studies of an improved solidification method of resins in cement are therefore performed by the utilities.

Some ion exchange resins, notably from decontamination work, contain organic acids that can adversely affect the solidification of the waste in cement. A method has been developed to incorporate these wastes into cement. It includes a pretreatment of the waste to destroy the acids followed by normal mixing with cement. Tests have been carried out on full scale (1 m³) samples with good results.

2.3.4 Ion exchange resins solidified in bitumen

The ion exchange resins are dried before they are solidified in bitumen or during the solidification. When they have been placed in a final repository, they will absorb water again with time. This process is very slow, since the bitumen impedes the water from penetrating into the ion exchange grains. After a long period of time, however, it must be assumed that the grains will be saturated with water. The swelling of the bitumenized waste could lead to damage of a barrier in the final repository. In their evaluation of the safety of SFR, the Swedish nuclear power inspectorate stated that such a situation is not acceptable.

Several studies have been performed to further clarify the swelling process. These studies include determination of swelling pressures for naked dried ion exchange resins as a function of the water uptake, and determination of water uptake rate in bitumenized resins. The measurements are complemented by a theoretical study. Some factors that affect the swelling are:

- type of resin and resin concentration
- drying temperature and time
- pretreatment
- residual water content
- absorbed radiation dose
- water quality

The measurements show that swelling pressures of 10 MPa or more can be obtained if the resins are hindered from swelling during water uptake.

The water uptake rate in bitumenized resins is primarily determined by the resin/bitumen ratio. Experiments indicate that if the

resin content is less than 50% the time needed to fill the 10% void space in a waste package is more than 1 000 years.

All measurements have been performed on samples prepared in the laboratory. However, tests are in progress with samples extruded from the solidification facilities at the nuclear power plants. Preliminary results indicate that the swelling is less for these samples.

Another question of interest for the final disposal of bitumenized waste is the possible attack of microbes. A literature survey has been done and experimental work on both aerobic and anaerobic degradation of bitumen is in progress.

2.3.5 Other treatment methods for ion exchange resins

In the annual report of 1981 the outlines of a new treatment method for ion exchange resins, called PILO, was described. In brief, it involves transferring the radioactivity from the organic ion exchange resins to inorganic ion exchangers (titanates and zeolites). The method affords the following advantages:

- * After heat treatment of the inorganic ion exchangers, a volume reduction of the waste by a factor of 5-10 is obtained compared to the untreated organic ion exchange resin.
- * The activity is bound in a matrix with very good long-term stability and leaching properties.

During 1982 the experimental work on the PILO-process was completed. An evaluation of the process has then been done to determine whether the PILO process entails such improvements with respect to safety and economy in comparison with currently applied conditioning methods that a full-scale adoption of the method in the Swedish nuclear power system is justified. The result of this evaluation was that, in spite of the reduced waste disposal costs, the overall system costs would be greater with the implementation of PILO in the Swedish system. (KBS TR 83-23) One important factor in the evaluation was the fact that PILO would replace already installed systems. Further development work on PILO has therefore been phased out from the KBS program.

For low-level resins from the condensate clean-up system incineration could be an alternative. Pyrolysis is therefore being studied by the Swedish utilities.

2.3.6 Quality control of reactor waste

Since the general outline of the final repository for reactor waste now has been determined, it is possible to specify quality

requirements on the reactor waste. A study with this purpose is running. The aim of the study is to identify the characteristics of the waste that determines the safety during the different steps of handling, storage and disposal and to determine the quality control and assurance program required.

2.3.7 Alfa-content in reactor waste

Although the radioactivity content of the reactor waste primarily consists of corrosion and fission products with a rather short half live (30 years or less), also traces of long-lived alfa-emitting radionuclides will appear. The two main contributions of alfa-activity in SFR will come from primary water clean-up resins and from fuel element crud that is released at the reception of fuel at CLAB. To get a basis for the evaluation of total alfa-content in SFR a series of measurements has been done in some Swedish reactors. These include analysis of reactor water and fuel scrapings. The results indicate that the short-lived ^{242}Cm gives most of the alfa-activity (90%) and that the content of long-lived alfa-emitters is normally less than 1GBq/reactor year.

2.3.8 Corrosion of steel in concrete

A potential means of off-setting the barrier function in the silo repository in SFR is the build-up of gas-pressure inside the silo. One mechanism for gas production that has been identified is hydrogen producing corrosion of the steel under anoxic conditions. The concrete in the silo guarantees that a high pH will be maintained around the steel in the repository (in waste and construction reinforcement) which leads to a very slow corrosion rate, if any. Experiments are in progress, which indicate corrosion rates around $1\mu\text{m}/\text{year}$.

2.4 **WASTE FROM THE DISMANTLING OF NUCLEAR POWER PLANTS**

Only minor work concerning waste from the dismantling of nuclear power plants has been done by KBS during 1983 in connection with cost calculations for the back end of the nuclear fuel cycle. - Practical experience with the handling and classification of waste from dismantling has been obtained by Studsvik Energiteknik in connection with the dismantling of the research reactor R1 in Stockholm.

Preliminary studies are in progress within the State Power Board regarding the future fate of the small PHWR Agesta which was decommissioned 1974 and since then been kept in protective storage.

3 MATERIALS

3.1 SPENT FUEL

Two high-level waste forms, spent fuel and vitrified waste from reprocessing, are currently being considered within the KBS studies. During 1983, the main thrust of activities has been on spent fuel. The studies on spent fuel characterization and leaching are made in close contact with groups in Canada and United States. A third workshop on the subject was organized in November 1983 in Boston (KBS TR 83- 76).

The experiments /3-1/ carried out for KBS at Studsvik during 1982-1983 have been made with segments of BWR fuel with a burnup of 41 MWD/kgU. Segments of fuel and cladding were cut from different zones along a well characterized fuel rod but with about the same burnup and duplicate tests were always performed using fuel from different zones.

A modified IAEA test was carried out in deionized water (pH 7.0), while all other tests employed synthetic groundwater (see Table 3-1). On completion of each leach period an aliquot of the leachant was filtered through a 1.5 - 2.0 mm membrane filter. The centrifugate was considered to contain the fraction in true solution and the fraction retained on the filter as a colloidal fraction.

Table 3-1. Composition of the synthetic groundwater, used for leaching of spent fuel.

Species	HCO_3^-	SiO_2	SO_4^{2-}	Cl^-	Ca^{2+}	Mg^{2+}	K^+	Na^+
ppm	123	12	9.6	70	18	4.3	3.9	65

pH: 8.0 - 8.2. Ionic strength: 0.0085.

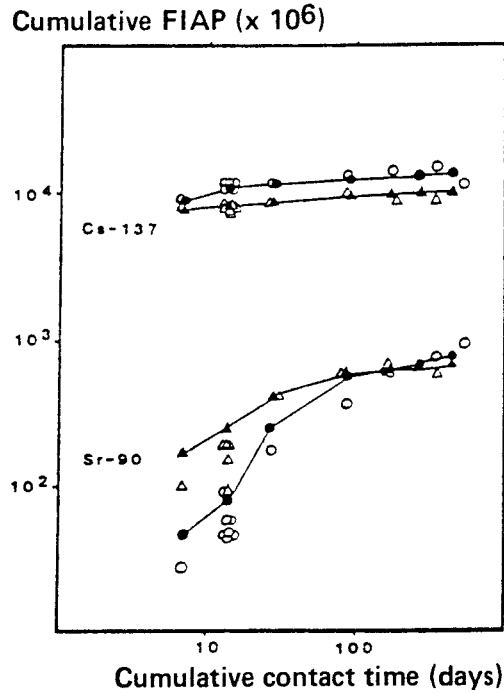


Figure 3-1. Comparison of results from sequential (IAEA-type) and static leaching in groundwater for duplicate specimens.

Open symbols: static tests.

Filled symbols: sequential tests.

FIAP denotes fraction of inventory in aqueous phase, i.e. sum of centrifugate and colloidal fraction.

Most of the experiments were performed under oxidizing conditions in order to provide benchmarking data and to permit comparison with other published results. However, some experiments were performed attempting to impose reducing conditions, which are expected to prevail in a repository.

3.1.1 Oxidizing conditions

As expected, about 1% of the Cs-137 inventory was rapidly leached out; most during the first 10 days. After this initial phase, the leaching decreased considerably and for longer periods of time, the leach rates were in the range $4 \cdot 10^{-7} \text{ d}^{-1}$ after 450 days. As is evident from Figure 3-1, there were initial differences between the specimens from different parts of the fuel rod. These differences had virtually disappeared after about 100 days of leaching. These initial differences, which are not reflected in corresponding leaching of uranium and actinides (see Figure 3-2), indicate selective leaching of Sr at least initially.

The leach rate for uranium was about $7 \cdot 10^{-8} \text{ d}^{-1}$ while for Plutonium and Curium values of $3 \cdot 10^{-9} \text{ d}^{-1}$ were found.

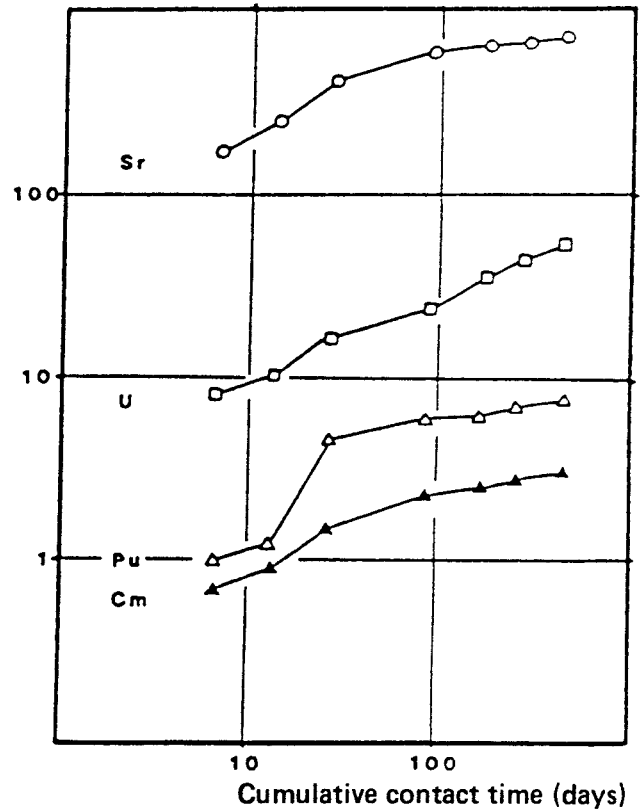
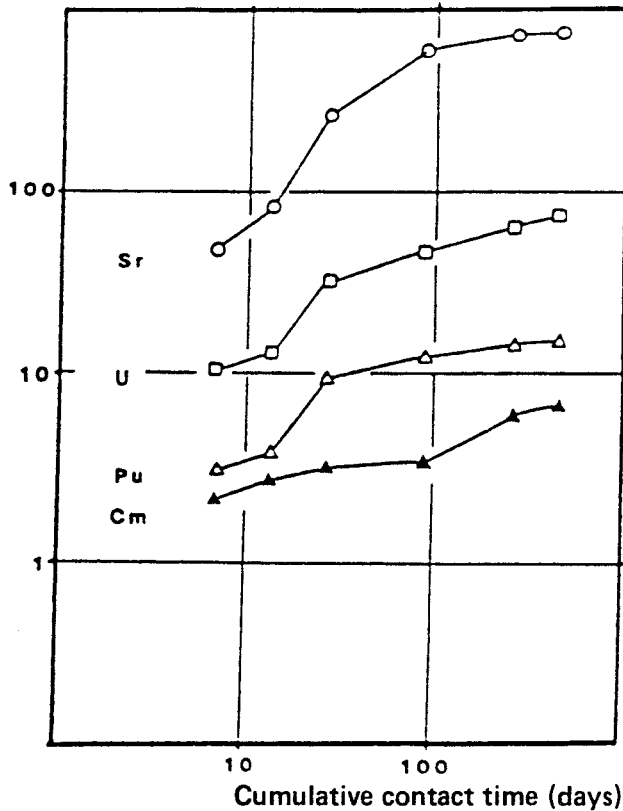
Cumulative FIAP (x 10⁶)

Figure 3-2. Sequential (IAEA-type) leaching results for groundwater. Duplicate specimens.

3.1.2 Solubility limitations

When the concentrations in the centrifugates are plotted as a function of contact time, it can be seen that uranium reaches an apparent saturation limit after about 10 days at a level of about 800 ppb (see Figure 3-3). This is in good agreement with the values calculated by Johnson /3-2/ based on data published by Paquette and Lemire /3-3/, but considerably lower than the concentrations conservatively estimated from the carbonate concentrations in the synthetic groundwater (123 ppm HCO_3^- should lead to 160 ppm $\text{UO}_2(\text{CO}_3)_3^{4-}$).

This discrepancy cannot be accounted for by absorption of uranium on the vessel walls. Normally, only small quantities were found when washing the vessels in decontamination solution, the maximum quantities being in the same order of magnitude as those fractions found in the aqueous phase. However, very slow reaction kinetics may be operative and cause this apparent saturation.

The results for the centrifugates of deionized water seem to indicate a saturation much lower, about 20-30 ppb. Complexing of uranium by the carbonate in the groundwater is the probable reason for these differences.

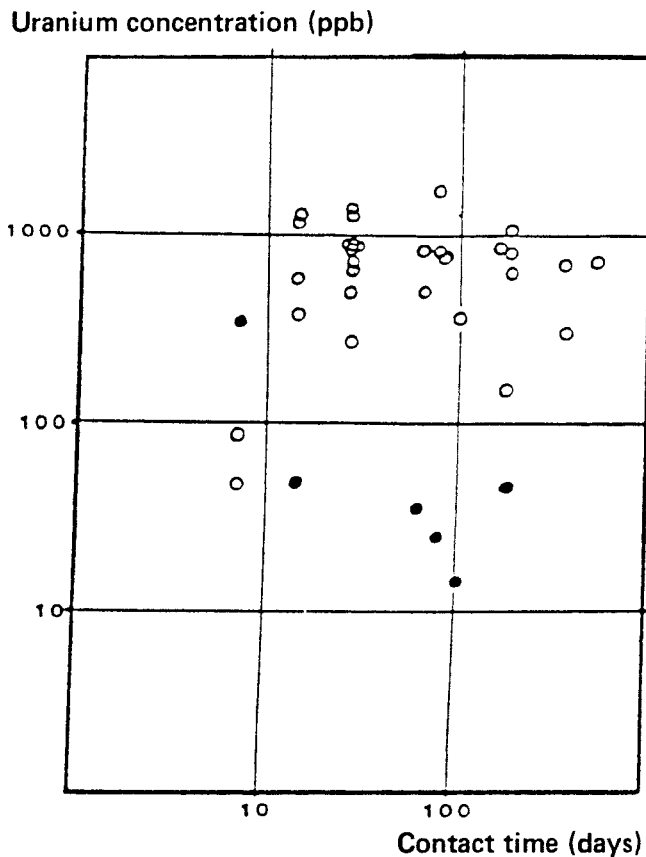


Figure 3-3. Concentrations of uranium found in leach solution centrifugate. Open symbols: groundwater. Filled symbols: deionized water.

3.1.3 Reducing conditions

Some experiments to impose reducing conditions have been performed using hydrogen gas in the presence of a palladium catalyst as reducing agent. These experiments must be regarded as preliminary method trials, but significantly lower leach rates were achieved. While for oxidizing conditions, centrifugate concentration of uranium of about 800 ppb were found, the values for reducing conditions represent results at the analytical detection limit, which is around 15 ppb.

3.2 WASTE GLASS

3.2.1 Simulated inactive glass

For nuclear waste glass studies, the results from 1983 refer to the phase of the ongoing program, i e studies of the effects of repository components interactions on the release for vitrified waste. Three glass compositions have been studied, ABS 39, 41 and 118. ABS 118 being a nonradioactive simulation of the waste

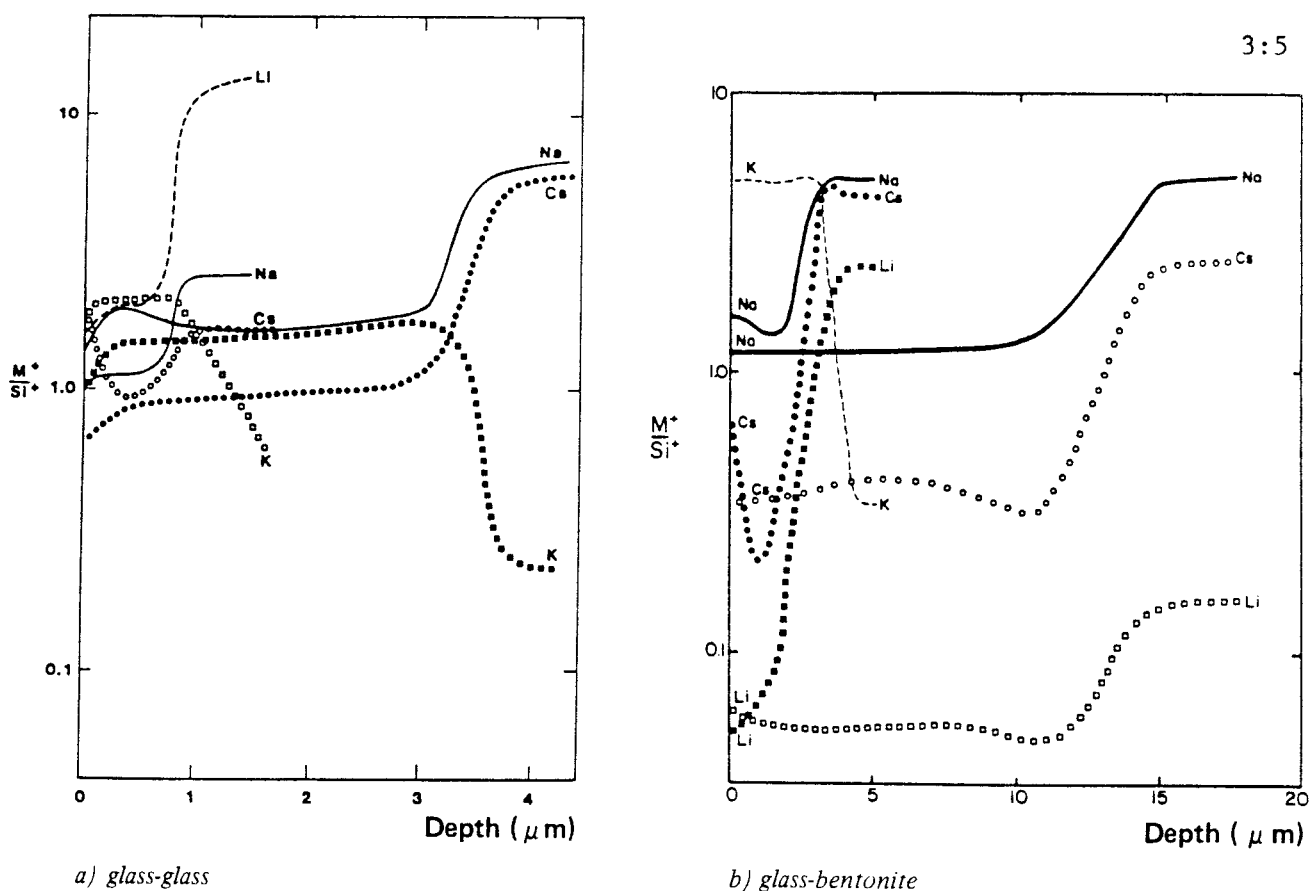


Figure 3-4. Alkali SIMS in-depth profiles from glass-glass interfaces (a) and glass-bentonite interfaces (b). The deeper profiles refer to glass ABS 39. The shallow profiles refer to glass ABS 41. Note the different scale in a) and b).

glass proposed by COGEMA for the UP3 vitrification plant in La Hague.

The investigations have included studies of the influence of bentonite, granite and stainless-steel corrosion products on the leaching of glass. Generally, the findings are that the presence of granite alone seems to lower the corrosion rate of the glasses, probably due to earlier occurring saturation. However, bentonite, compacted as well as uncompact and even water equilibrated with bentonite proved to strongly enhance the corrosion of a nuclear waste glass /3-4/.

The ongoing Stripa burial experiments using the glass compositions ABS 39 and ABS 41 have been evaluated in exposure times up to 12 months /3-5, 3-6/. The results confirm the findings from laboratory experiments, i.e. the contact with bentonite produces a rapid corrosion attack extending about 2-3 times deeper than the corresponding corrosion depth for a glass-glass interface (see the SIMS alkali profiles shown in Figure 3-4). However, the results indicate that the initially high corrosion rates produced by the bentonite interface is rapidly slowed down after the corrosion layer has developed and thereafter approach the corrosion rates of glass-glass and glass-granite interfaces (see Figure 3-5).

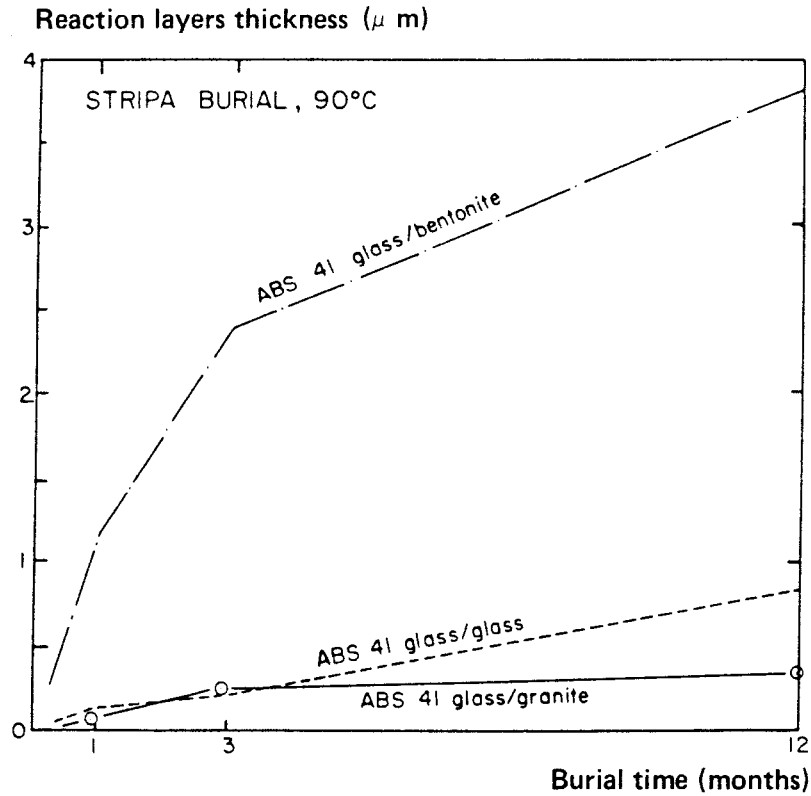


Figure 3-5. Reaction layers depth as a function of time for different interfaces.

3.2.2 JSS-project

The JSS-project is a joint project between SKBF/KBS, NAGRA (Switzerland) and CRIEPI (Japan).

The phase I of the JSS-project has been completed and the final report for that phase is currently being prepared. The general finding is that the radioactive glass behaves similarly to the non-radioactive simulation, ABS 118, and that no effects of radiation of the corrosion behaviour has been found so far.

Prolonging the project with a phase IV is currently discussed, but the details of the program are still to be decided.

3.3 CANISTER MATERIALS

3.3.1 Copper

The main thrust during 1983 has been directed towards copper as canister material. Most of the studies on copper are reported in more detail in the KBS-3 report and will therefore only be briefly reviewed here.

Copper corrosion

The assessment of the corrosion of a copper canister in the repository environment has been updated (KBS TR 83-24). Since copper is thermodynamically stable in pure water, the corrosion rate is controlled by the transport of corrodants from the surroundings. Initially, oxygen trapped in the deposition holes and the tunnels will be the major corrodant, but for longer periods of time, the corrosion will be controlled by the supply of sulphide from various sources. The total uniform corrosion attack on a canister can be seen in Table 3-2.

Table 3-2. Depth of uniform attack (mm) on a copper canister caused by reactants supplied from the outside as a function of exposure time.

Reactant	Exposure time, years			
	10^3	10^4	10^5	10^6
<u>Oxygen</u>				
Initially in deposition hole	0.04	0.04	0.04	0.04
From tunnel without vivianite addition	0.03	0.03	0.03	0.03
From groundwater via fractures	0.00	0.00	0.00	0.02
<u>Sulphide</u>				
Initially in deposition hole with 0.02% S in bentonite, incl microbially formed sulphide	0.05	0.05	0.05	0.05
From tunnel with 0.02% S ²⁻ in bentonite, incl microbially formed sulphide	0.00	0.00	0.04	0.37
From groundwater via fractures, incl microbially formed sulphide	0.00	0.00	0.02	0.17
Total, mm	0.12	0.12	0.18	0.68

In addition to a uniform attack, several localized corrosion phenomena may occur. In particular, pitting and stress corrosion cracking have been studied.

In order to better elucidate the pitting corrosion in copper, archaeological copper and bronze objects have been studied and the pitting factor measured (KBS TR 83-05). Generally, low pitting factors were found (≤ 5) supporting the conclusion that the pitting factor decreases with time (KBS TR 83-24). A study of some older lighting conductor plates also revealed very low pitting factors (KBS TR 83-24).

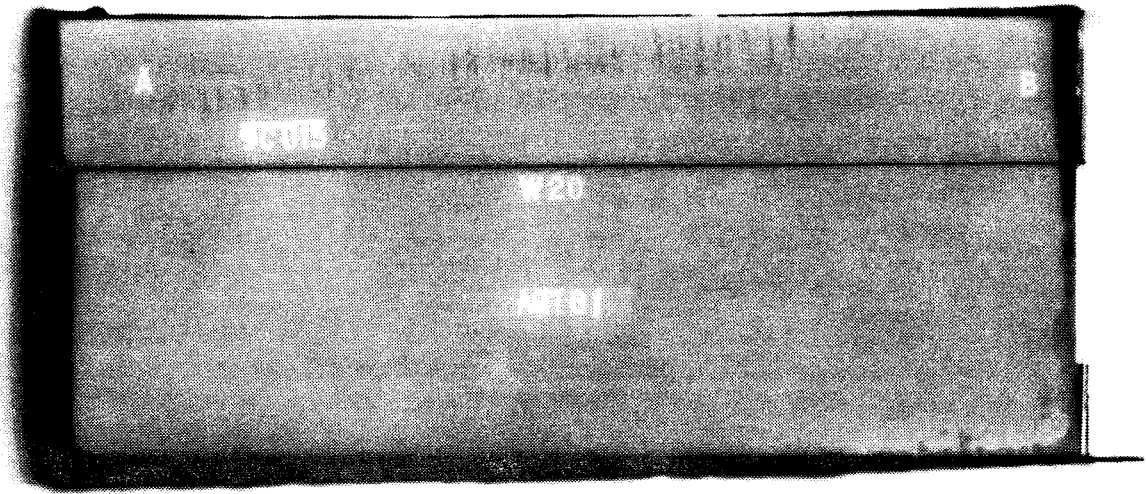
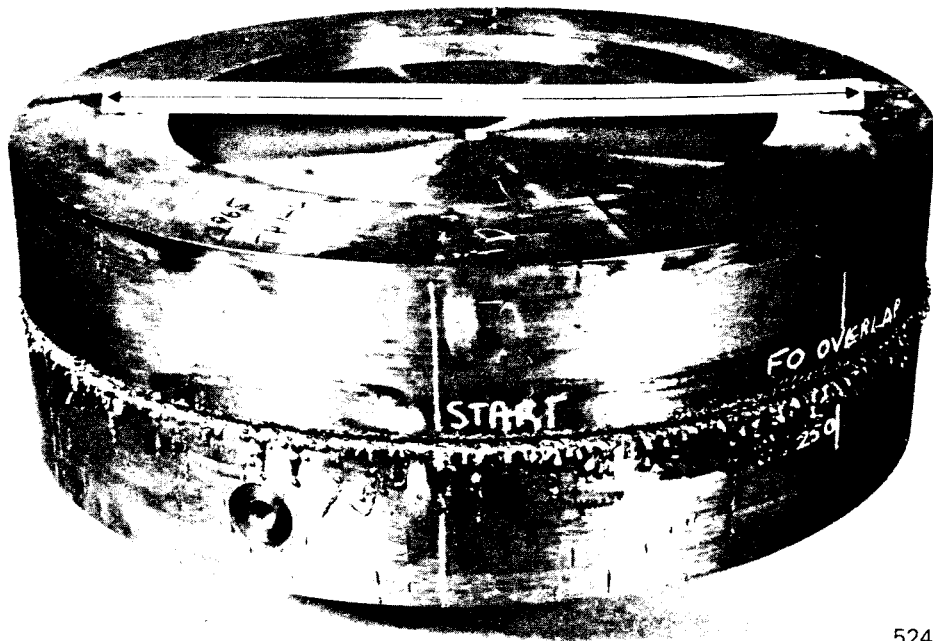


Figure 3-6. X-ray radiogram of a defect-free electron beam welded section in 10 cm thick copper. In the 5 cm thick backing (section A-B) some root defects can be seen.

The susceptibility of copper to stress corrosion cracking was studied using the slow strain rate testing in various environments. In the tests, both aerobic and anaerobic conditions were used and the results indicated lack of susceptibility to stress corrosion for the groundwater compositions likely to prevail in the repository. However, as expected nitrite contamination of the groundwater was found to promote stress corrosion, but only at concentrations exceeding 69 ppm (KBS TR 83-06).

Welding of copper

A feasibility study of electron beam welding of thick copper has been completed and show that full penetration of 100 mm thick OFHC copper can be achieved at a power level of 75 kW (KBS TR 83-25). For steady state conditions, a defect free weld can be made, see Figure 3-6. However, in ending the weld (fade out) it is still not possible to completely eliminate root defects. A semi-full-scale lid weld has also been performed successfully, see Figure 3-7).



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Fig. 3-7. General view of simulated lid assembly as-welded.

3.3.2 Titanium

Titanium has been proposed as canister material and it is the major candidate material among passivated metals. A study of the kinetics and structure of the passive film generated on titanium in aerobic and anaerobic conditions has been completed /3-7/. The study shows an amorphous oxide film with a thickness that grows linearly with the logarithm of time as expected for these types of metals. The growth rate is independent of whether oxygen is present in the water or not (see Figure 3-8). The growth rate of the oxide film formed in contact with bentonite is found to lie on the same curve for exposure times up to two years. The regeneration rate of the film is low; in three months a layer of about 1Å of the titanium metal has left the specimens.

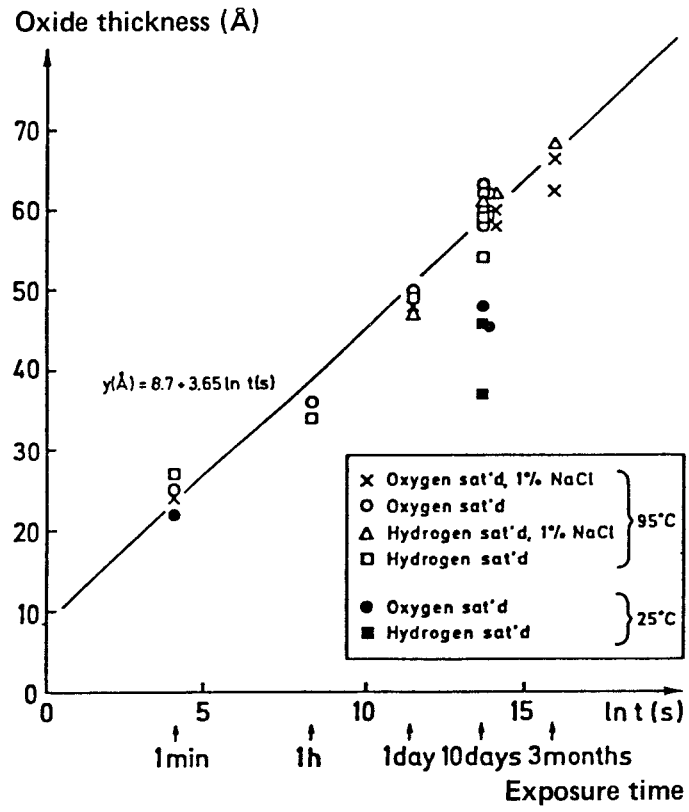


Figure 3-8. Oxide layer thickness versus exposure time for electropolished Ti exposed to water of various compositions.

4 ENGINEERED BARRIERS, DESIGN AND TECHNOLOGY

4.1 CANISTERS

Two alternative designs of copper canisters have been further studied; one with filling of lead and sealed by use of electron beam welding and the other with filling of copper powder which is solidified by use of hot isostatic pressing, the HIP method.

4.1.1 Welded copper canister

As mentioned in Chapter 3 electron beam welding of thick copper has been tested on a canister lid in full scale. The tests show the feasibility of welding of more than 100 mm thick copper (KBS TR 83-25).

Two methods for pouring molten lead into a copper cylinder have been tested. A cylinder with a simulated BWR fuel bundle with reduced length, (2 m) was used. In both cases the cylinders were filled from the bottom and up through a steel tube. In one case the contraction during cooling was compensated for by a second filling, in the other case by artificial cooling of the bottom. The test results show that the open space in a canister can be filled up by lead to more than 98 per cent, giving a final void of less than 2%.

4.1.2 HIP canister

Two top parts of canisters in about full-scale have been fabricated for demonstration of the technical feasibility of embedding each fuel rod in a canister with solid copper by using hot isostatic pressing (HIP). The canister diameter was 610 mm before pressing, had a length of about 2 m and a total weight of 4 300 kg, see Figure 4-1. In the HIP process pressures up to 150 MPa and temperatures up to 500°C were used. Testing of the solid cop-



Figure 4-1. A HIPed canister with seven shortened simulated fuel bundles of BWR type embedded in copper. Dimensions $\text{\O} 600 \times 1800$ mm. Weight 4300 kg.

per from pressed powder have shown values of strength and ductility corresponding to copper in annealed condition.

The HIP process, caused a shrinkage of 4% on the diameter and 1% on the length.

4.2 FACILITY DESIGN STUDIES

The main features of the KBS-3 repository concepts are shown in Figure 4-2.

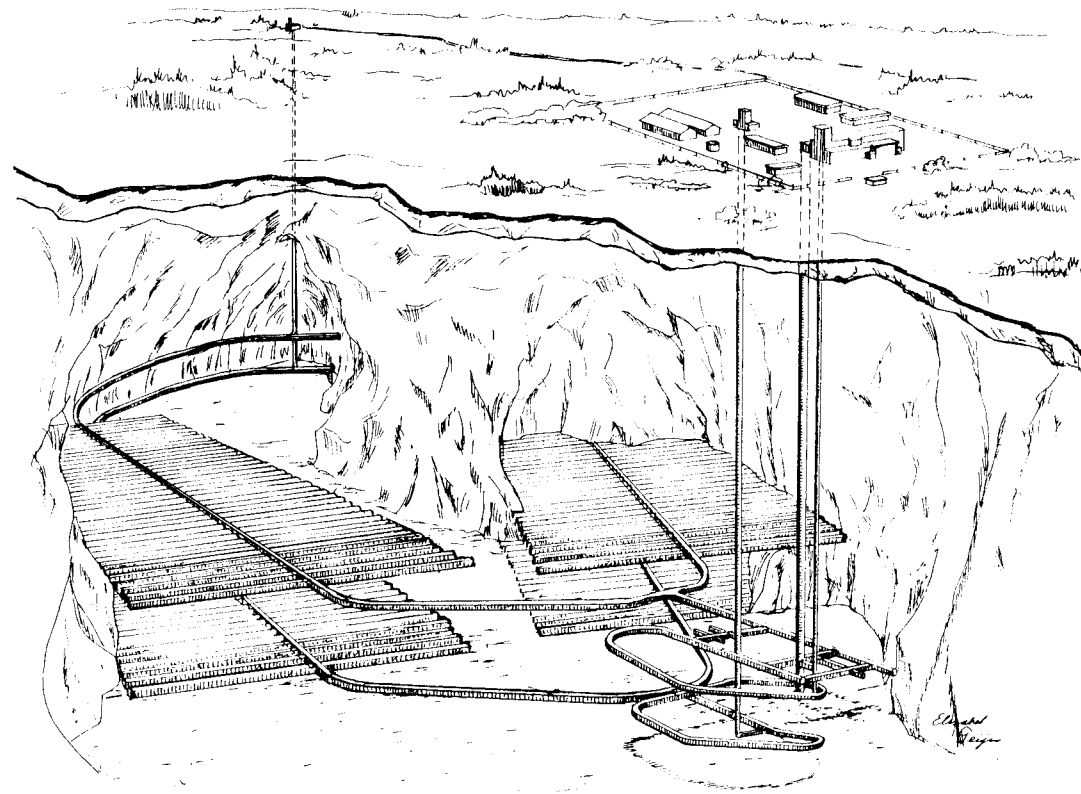
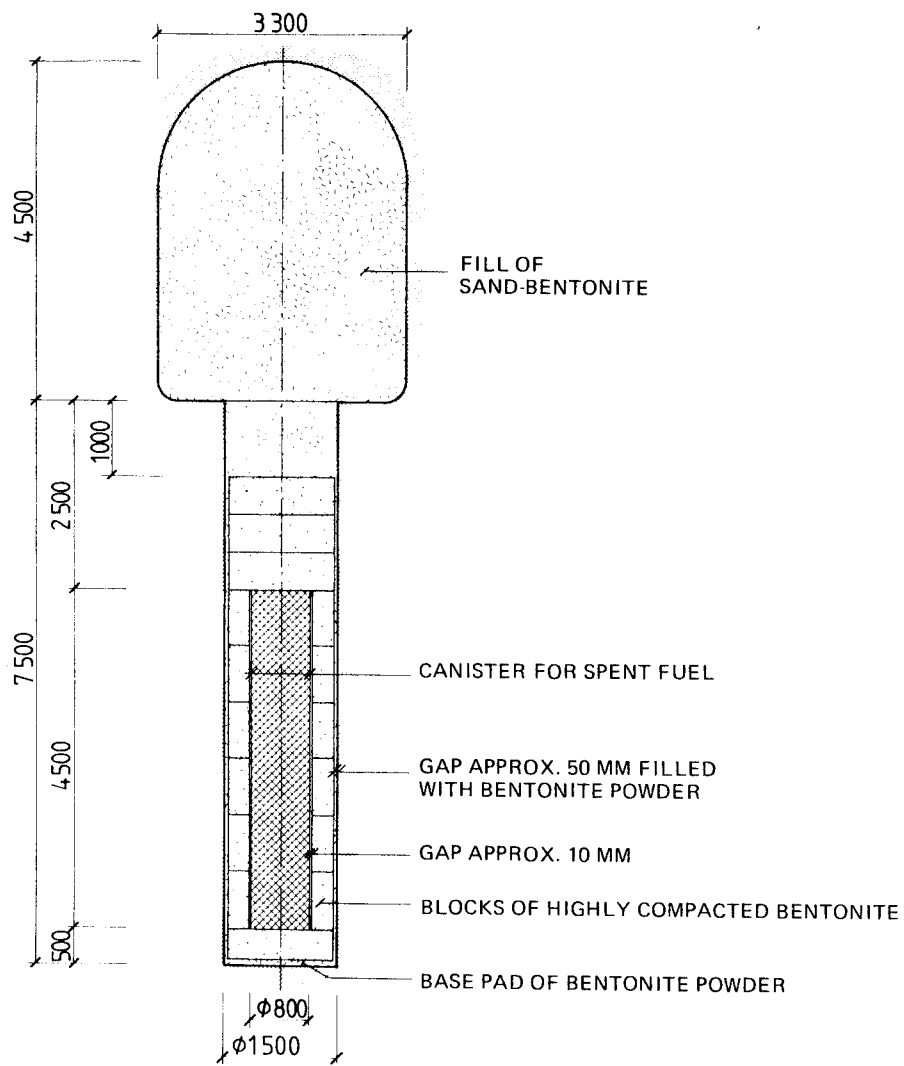


Figure 4-2. Deposition hole with canister and buffer material and with backfill in storage tunnel (left). Tunnels and shafts in the final repository for spent fuel (right)

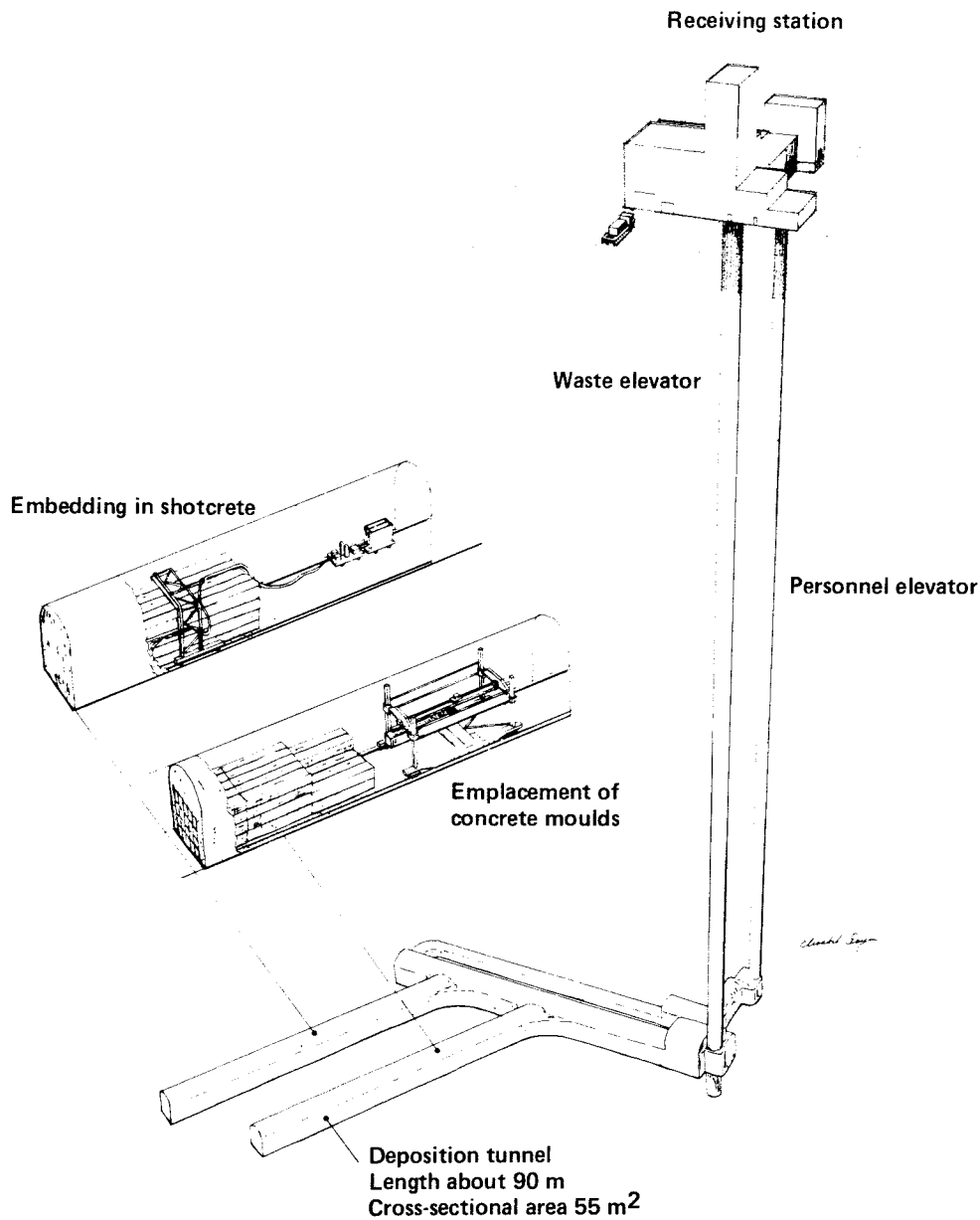


Figure 4-3. Final repository for fuel boxes etc.

Some further calculations of the temperatures in the repository have been made (KBS TR 83-22). Studies have also been performed of the design of a separate repository for BWR-boxes, control rods etc (KBS TR 83-20), see Figure 4-3.

4.3 CLAY BARRIERS

4.3.1 General

In the concepts for final repositories for spent fuel, and for reactor waste, SFR (see Chapter 9) bentonite based clay-mixtures

are used as components in the multibarrier system. The main functional requirements of these buffer- and backfill materials are

- * low hydraulic conductivity
- * sufficient bearing capacity
- * sufficient ductility to eliminate negative consequences of possible rock movements
- * swelling potential to fill up voids
- * sufficient heat conductivity
- * chemical and physical stability

During 1983 the studies has focused on the physico-chemical properties of smectite-rich clays, the rheological behaviour of highly compacted bentonite and the chemical and physical stability of bentonite gels.

Highly compacted bentonite has been studied in a realistic environment in the Stripa project, see Chapter 10.

Methods for application of bentonites with lower densities suitable for use in SFR, see Chapter 9, have been tested.

4.3.2 Rheological and physical properties of bentonite gels

The stress/strain behaviour of dense bentonite was measured in laboratory experiments in order to model the creep behaviour with undrained and drained conditions and at elevated temperatures. Nuclear magnetic resonance technique (NMR) was used for measurements of the spin-spin coherence time (T_2) which was found to be very short for a water content in the interval 25-35%, which points to highly ordered water lattices. The main objective for further research with use of this technique is to improve the physical model for water/smectite interaction and to use this technique in connection with laboratory experiments for studies of chemical alterations at higher temperatures. Viscometer tests and pinhole experiments were performed in laboratory (KBS TR 83-04).

Theoretical models for clay gel migration into rock joints were used and the risk of erosion by flowing groundwater was estimated by comparing clay particle bond strength evaluated from viscometer tests and theoretically derived drag forces (KBS TR 83-04).

4.3.3 Chemical stability of smectite minerals

Natural analogues on smectite alteration under known geological, temperature and pressure conditions may give information of smectite/illite conversion over long time periods.

Studies - sampling and mineral analysis - of clay formations at Kinnekulle and Höganäs have been performed. The results indicate that a possible cementation mechanism, i e quartz precipitation, may be associated with the smectite/illite conversion (KBS TR 83-16).

A workshop on the long term stability of smectite minerals was organized in cooperation with DOE in Washington D.C. in December, 1983.

4.3.4 Sealing of boreholes, tunnels and shafts

Tests of highly compacted bentonite as sealing material in boreholes and shaft- and tunnel plugs, have started in Stripa, see Chapter 10.

5 CHEMISTRY

5.1 GENERAL

The chemistry program comprises the study of chemical conditions that influence the degradation of the canister material, the dissolution of the waste matrix and the transport of radionuclides in the buffer and the rock. The studies are divided into the following four categories.

- o Groundwater chemistry investigations, including groundwater characterization and efforts to understand the variability due to natural circumstances or induced by the near-field reactions in the repository.
- o Radionuclide chemistry comprising the build-up of a firm chemical data base on the behaviour of radionuclides in the anticipated environment and correlated radiolytical effects.
- o Model development regarding canister degradation, the release of radionuclides to the rock-groundwater environment and the transport and dispersion of released radionuclides in that environment.
- o In-situ tests and natural analogue studies intended to bridge the gap between on one hand theoretical and short-term laboratory studies and on the other hand processes in a realistic environment and during a realistic, geological time frame.

5.2 GROUNDWATER CHEMISTRY

5.2.1 Groundwater analyses and characterization

The year 1983 has been an intense period of site investigations and a large number of groundwater samples have been collected

and analysed. The results from the areas Gideå (KBS TR 83-17), Fjällveden (KBS TR 83-19), Svartboberget (KBS TR 83-41) and Kam-lunge (KBS TR 83-70) have been compiled and reported.

A general characterization of groundwater at depth on the basis of these and related results has been reported (KBS TR 83-59). The important and interrelated parameters pH and carbonate content have also been dealt with and reported (KBS TR 82-25). A further evaluation of groundwater composition including advanced geochemical model calculations is under way.

The composition of groundwater at depth in granitic rock is generally within the limits given in Table 5-1. The irrational occurrence of highly saline water is a notable deviation, the origin of which is being investigated. Interpretations so far favour the rock itself as a major source rather than a marine origin. The implications of raised salinity for the copper canisters and radionuclide chemistry have been evaluated in the KBS-3 study.

Table 5-1. Normal composition of deep granitic groundwater in Sweden (concentrations in mg/l).

pH	7 - 9	Ca ²⁺	10 - 40
Eh, V	0 - (-0.45)	Mg ²⁺	2 - 10
HCO ₃ ⁻	90 - 275	Na ⁺	10 - 100
SO ₄ ²⁻	0.5 - 15	K ⁺	1 - 5
HPO ₄ ²⁻	0.01 - 0.2	Fe ²⁺	0.02 - 5
NO ₃ ⁻	0.01 - 0.05	Mn ²⁺	0.1 - 0.5
F ⁻	0.5 - 4	NH ₄ ⁺	0.05 - 0.2
Cl ⁻	4 - 15	SiO ₂ (tot)	3 - 14
HS ⁻	0 - 0.5	TOC ^a	1 - 8

a Total content of organic carbon.

The organic content of the groundwater have been further identified as humic and fulvic acids which like the carbonate ions are important as potential complex forming agents for the actinides.

Data on natural radioactive and stable isotopes in the groundwater, which could be helpful to tell the history of the water, have been collected. However, evaluations have not yet been successful due to interpretation problems identified in the course of the investigations /5-1 and 5-2/. The intriguing question of groundwater history will be further studied. This is also an important part of the studies performed by the international experts of the hydrochemistry group within the Stripa project.

5.2.2 Redox conditions

The presence of iron(II) in minerals is expected to cause reducing conditions at depth; ensuring the stability of copper canisters and giving a low solubility and low mobility to most of the radionuclides of interest (TR 83-38). The direct measurement of redox potentials in groundwater is generally recognized as being a difficult task /5-3/.

Special equipment has been developed for the direct electrochemical measurement of Eh in the field and great care has been taken to sample and analyse the redox sensitive groundwater components such as iron, manganese and sulphide, dissolved oxygen and uranium. The results have been evaluated and reported together with supporting simulating laboratory experiments (KBS TR 83-40). The measurements clearly shows the anticipated reducing conditions in the deep Swedish crystalline bedrock.

In parallel a new generation of equipment is being developed where the redox potential is measured in situ. This outfit is being integrated with a mobile field laboratory, which is under construction.

5.2.3 Fracture minerals and thermal effects

A typical groundwater composition is obtained in a process where infiltrating surface water, low in dissolved solids, takes up carbon dioxide from the biological activity in the root zone and reacts further with the minerals on the way down to deeper levels. The fracture minerals of Gideå have been carefully investigated which has validated the simplified description above and also shown evidence of early hydrothermal events (KBS TR 83-74).

The presence of a high level waste repository will create a thermal pulse in the surrounding rock. Geochemical model calculations of the effect of this pulse on the chemistry of groundwater and minerals are performed with Gideå as a typical area. This work is still going on and preliminary results (KBS TR 83-74) show a good potential of the approach used.

5.3 RADIONUCLIDE CHEMISTRY

5.3.1 Thermodynamic properties

Basic chemical data on the solubility, complex formation and redox properties of actinides and technetium under repository and ambient groundwater conditions have been compiled and evaluated in the course of the KBS-3 study (KBS TR 83-02, 83-35 and 83-60). Some of these data, especially on uranium, have been generated within the program /5-4/.

Coprecipitation is a limiting mechanism for the release of radionuclides from the repository. This has been studied for the precipitation of plutonium with uranium (KBS TR 83-02, appendix 2).

5.3.2 Colloides and organic complexes

The formation, stability and mobility of actinide colloides formed by precipitation or sorption on inactive particles have been studied (KBS TR 83-08). The sorption phenomena will be studied further.

Effort has also been directed to the evaluation of natural organic substances as potential carriers of released radionuclides (KBS TR 83-08 and 83-09). Experiments with and without organic complex-forming agents show that the sorption and retardation of the nuclides may be lowered by as much as one order of magnitude by the presence of these substances. (KBS TR 83-08, see also Figure 5-1).

5.3.3 Sorption and diffusion

The sorption of radionuclides on the mineral surfaces is an important retention mechanism. Data on the sorption of fission products (KBS TR 83-07 and 83-63) and actinides (KBS TR 82-21) have been compiled for the KBS-3 study. A considerable quantity of this information has been experimentally obtained within the program (KBS TR 82-21 and 83-63).

The importance of water phase conditions (KBS TR 83-61) and mineral surface properties (KBS TR 83-64) has been carefully investigated experimentally in a rather unique effort. These are important steps in the direction of a deeper understanding and theoretical modelling of the sorption phenomenon.

Diffusion of radionuclides into the micropores of the rock matrix will considerably increase the available mineral surface for sorption and thereby increase the capacity of the rock to retain radionuclides (see Figure 5-2). This effect has been demonstrat-

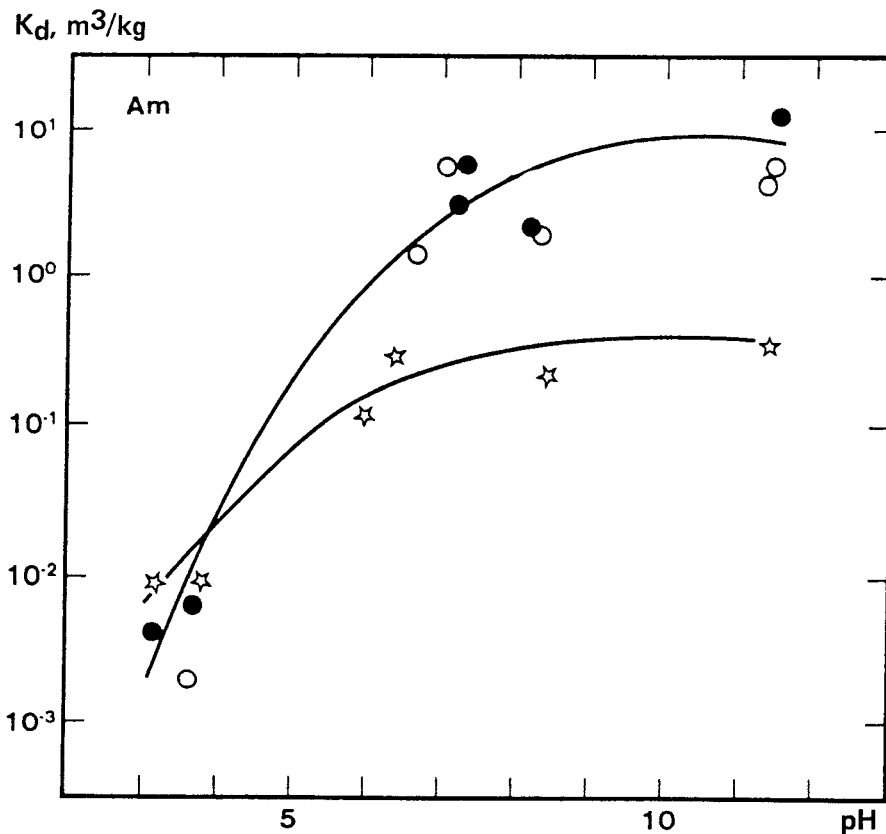


Figure 5-1. The distribution coefficient for americium as a function of pH in the granite-groundwater system in the presence of a strongly complexing humic acid, ○ without humus, ● 0,1 mg/l humus, ☆10 mg/l humus.

ed by laboratory experiments (KBS TR 82-12, 82-26 and 83-15) field experiments in Stripa (KBS TR 82-08, and 83-39), natural analogue investigations (KBS TR 82-15) and theoretical studies (KBS TR 83-37). This approach has also been used in the interpretation of in-situ migration experiments (KBS TR 83-38). The "into the rock matrix" diffusion concept has been an integrated part of the migration models used in the KBS-3 study (KBS TR 82-05).

Diffusion of different species through the bentonite buffer has been studied as a base for the evaluation of chemical transport in the near-field. The experimental studies comprise radionuclides (KBS TR 83-34) and inactive species such as colloides, organic substances, sulphide ions and hydrogen (KBS TR 82-17 and 83-71). The involved mechanisms have been discussed theoretically (KBS TR 82-27 and 83-37).

It should be noted that little emphasis has so far been placed on the retention of radionuclides in the buffer. The near-field transport of inactive species on the other hand determines the canister service life and the over all near-field performance.

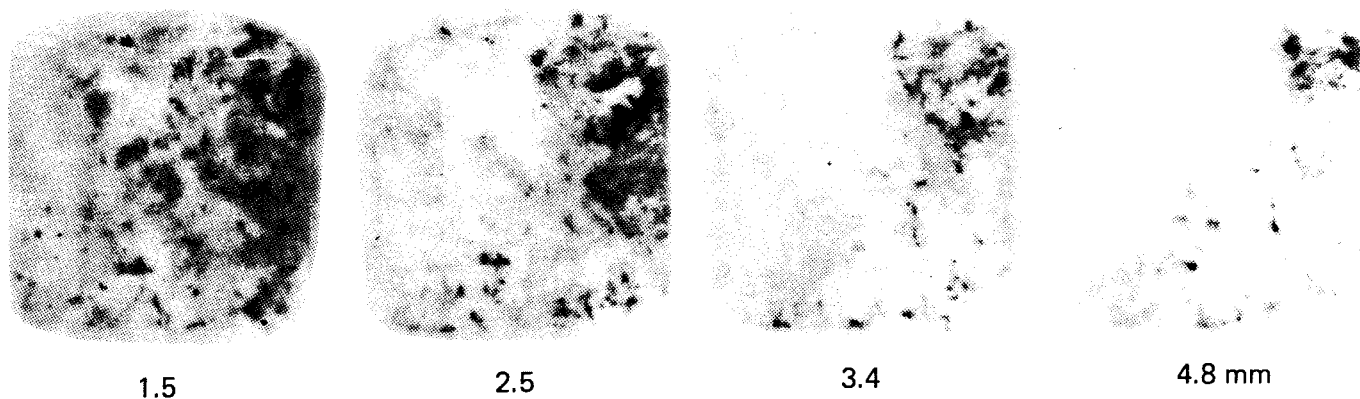


Figure 5-2. Autoradiographs showing the diffusion of ^{137}Cs into calcite-covered granite. Penetration depth given in mm. The thickness of the calcite layer is 1.5-2 mm. The sample containing a natural fracture with calcite-covered surfaces has been taken from a depth of 358 m in Finnsjön. The fracture surface has been contacted with the cesium solution for about 5 months.

5.3.4 Radiolysis

The effect of radiolysis from high level waste can be minimized during a long period of time by the shielding effect of the canister (KBS TR 82-02). However, if the canister in a far future is penetrated the alpha radiolysis may be of significant importance (KBS TR 82-18). Theoretical calculations supported by experiments with radiolysis of water in bentonite clay (KBS TR 83-27) have been performed, see figure 5-3.

The possible effects of alpha radiolysis on spent fuel dissolution (KBS TR 83-02) and on redox conditions in the near-field environment have been evaluated (see 5.4.1).

5.4 CHEMICAL TRANSPORT MODELS

5.4.1 Near-field transport models

The near-field includes the buffer and the first few meters of selected good rock around a deposition hole. Models have been developed which makes it possible to calculate the inward transport of corrodants to the canister and the outward migration of released radionuclides and radiolysis products (KBS TR 82-24).

Radiolysis of water in contact with exposed spent fuel will produce oxidizing substances and hydrogen. The hydrogen gas will escape easier by diffusion and thus leaving behind an oxidizing environment near the canister. The development of these conditions have been predicted by near-field transport models based on radiolysis calculations (KBS TR 82-16, 83-67 and 83-68).

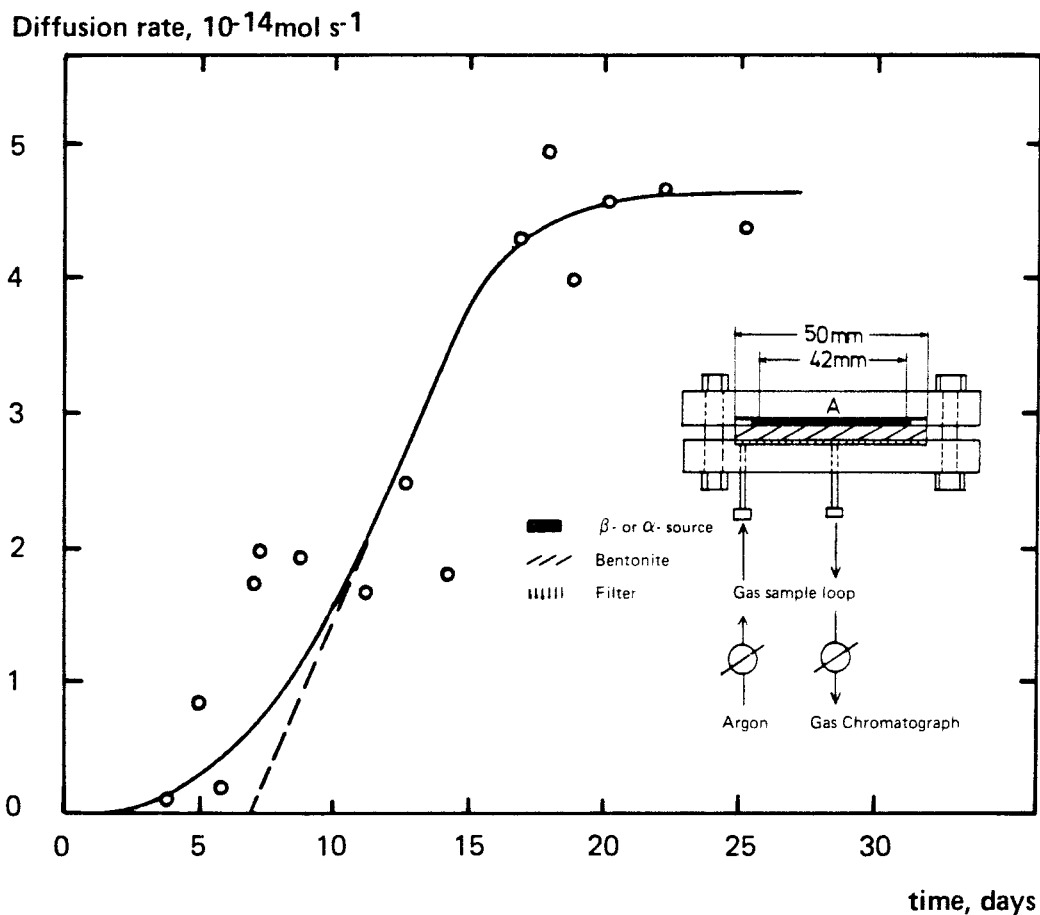


Figure 5-3. Hydrogen diffusion out of bentonite irradiated with a $200 \text{mCi } ^{147}\text{Pm}$ β -source. Dose rate 6 rad/s .

The released radionuclides will have to pass from an oxidizing near-field into the ambient reducing rock-groundwater environment which leads to important changes in the actinide and technetium chemistry. These phenomena have been modelled in order to get a source-term for the far-field migration models (KBS TR 83-66).

5.4.2 Far-field migration of radionuclides

When the radionuclides once have arrived into the far-field the disturbances from the repository (the near-field) are judged to be of minor importance and chemically the radionuclides will be transported by the normal reducing groundwater at a very low concentration. Numerous experiments have been performed to quantify the sorption and diffusion of the radionuclides in granitic

rock. Models have been developed which, based on the experimental data for chemical retention, calculates the dispersal of radionuclides in the rock.

The retention of radionuclides during transport with the groundwater in fracture zones have not been taken credit for in the KBS-3 study. These possible effects will be an important issue in further studies (KBS TR 83-69).

5.5 IN-SITU TESTS AND NATURAL ANALOGUES

5.5.1 In-situ tests

In-situ tests are logical steps in scaling up laboratory experiments which describe the radionuclide transport in the rock. As an introduction to such studies natural fractures are overcored and taken to the laboratory for fracture migration experiments. A number of such tests have been performed (KBS TR 83-01).

The use of nonsorbing tracers in crosshole tests to characterize the hydraulic conditions in the field is becoming a fairly well established technique. When it comes even to moderately sorbing species like strontium and cesium ions the studies become difficult due to the long observation times needed. Quite a lot of experience has been gained, however, in the following field experiments.

Studsvik

Migration experiments with radioisotopes of iodine, strontium and cesium have been performed in Studsvik (KBS TR 83-18), see also Figure 5-4.

Finnsjön

Inactive strontium and cesium have been used in field experiments in Finnsjön. The results have recently been reported but not yet published.

Stripa

An extensive program of migration experiments is being carried out in the Stripa Mine as a part of an international OECD-NEA Project. A large number of tracers, sorbing and nonsorbing, have been used (see Chapter 10).

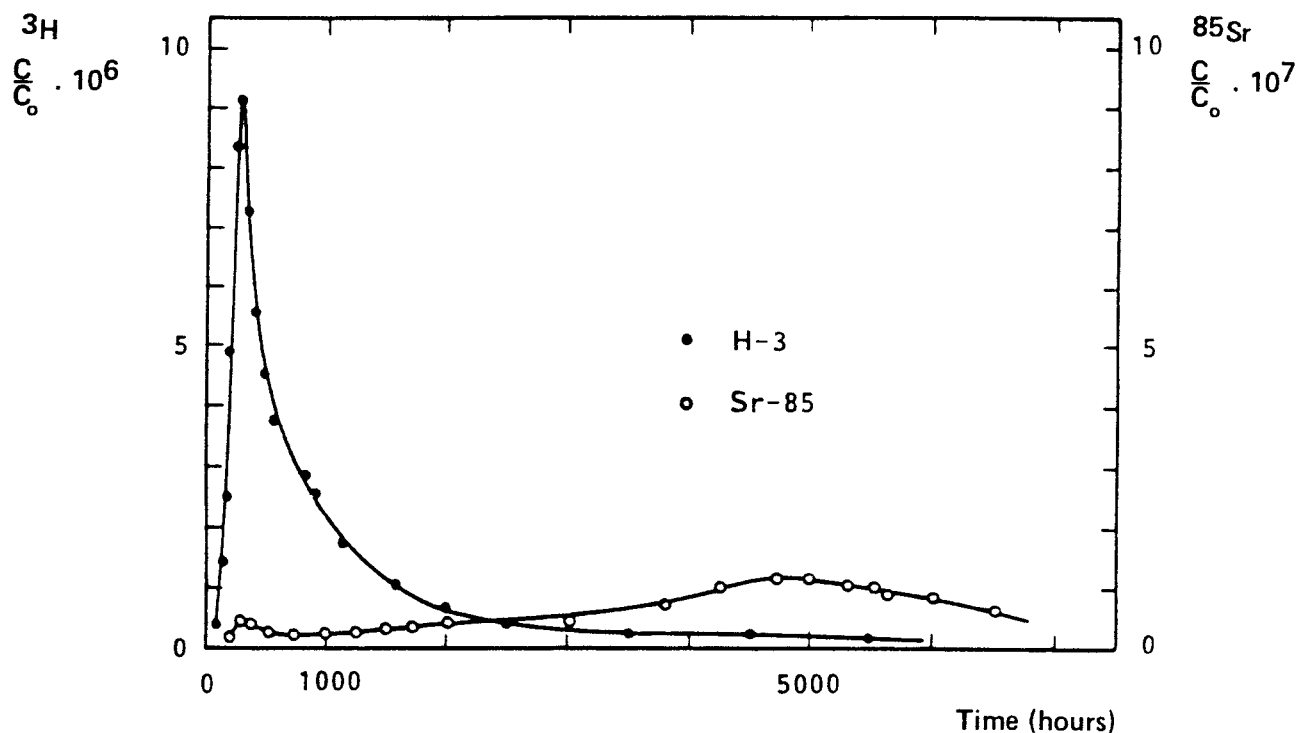


Figure 5-4. Field migration experiment in Studsvik. Break-through curves for H-3 and Sr-85. The flowpath consists of interconnected fractures at about 100m depth. Injection and sampling points are 12 m apart.

5.5.2 Natural analogue studies

The study of natural analogues to the migration of radionuclides from a repository is the only way to get "experimental" data from a realistic time period. To find analogues which are similar enough to the relevant problem or possible to document well enough is therefore an important but also a difficult task. During 1983 KBS has supported the following activities which are regarded as promising.

The Oklo phenomenon

The natural reactor in Oklo in Gabon had previously been carefully investigated in a number of studies /5-5 and 5-6/. KBS has supported a further evaluation of the Oklo phenomenon from the radiolysis point of view (KBS TR 83-10). Further studies are being planned.

Morro do Ferro

In cooperation with US, Swiss and Brazilian organizations KBS supports studies of Morro do Ferro - a concentrated thorium mineralization in Brazil. It is a good analogue to the dispersion of tetravalent actinides in the environment /5-7/.

Uranium mobility

The distribution of uranium in granitic rock (where it is a common constituent) or around uranium mineralizations can be used as an indicator of how uranium and other actinides will move in the rock-groundwater environment. Redox conditions are decisive which is clearly reflected in the solubility and uranium isotope distribution. These parameters are therefore regularly measured on groundwater and mineral samples from the investigation sites. A study has also been performed on a uranium mineralisation indicating diffusion of uranium into the adjacent rock matrix (KBS TR 82-15).

6 GEOSCIENCE

The activities within the field of geoscience are divided into two main parts:

- Site selection studies, i.e. investigation of the Swedish bed-rock in order to find sites suitable for final disposal of spent nuclear fuel.
- Research and development

6.1 SITE SELECTION STUDIES

The overall time-schedule for the site selection studies is shown in Figure 6-1. In total some 10-15 sites will be investigated by surface and borehole observations following a defined standard program /6-1/. Two or three of those sites will be selected for detailed studies around year 1990 and the recommendation for a specific site is expected to be given around year 2000.

The major rock types studied so far are granite, gneiss and to some extent gabbro. Further studies of gabbroic rock will be initiated.

Figure 6-2 shows the location of sites investigated by surface and borehole investigation and Table 6-1 summarizes the rock types and overall evaluation at the sites.

Below a short summary is given of results obtained during 1983.

6.1.1 Fjällveden

15 core boreholes have been drilled to a maximum depth of 700 m at the site. The total length of the core boreholes is about 7 600 m.

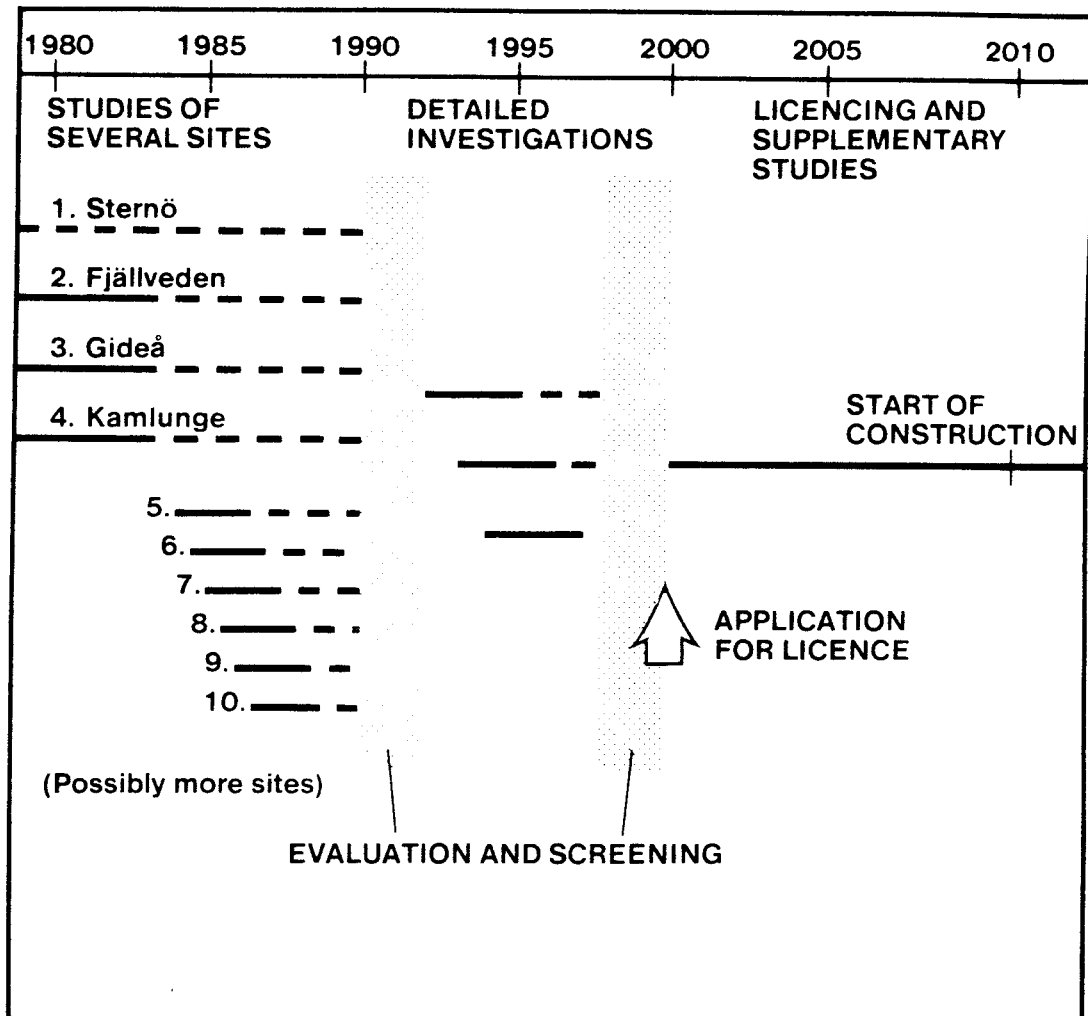


Figure 6-1. Time-schedule for site selection studies.

The site is characterized by a flat topography (KBS TR 83-52). The rock mass, which consists mainly of veined gneiss, generally possesses low hydraulic conductivity, which also applies to the local fracture zones. Approximately 3% of the rock mass consists of vertically oriented layers of gneissic granite with elevated hydraulic conductivity. Figure 6-3 shows the obtained hydraulic conductivity in the different rock types.

The groundwater flow at repository depth has been calculated to be between 0.01 and 0.05 l/(m² · year), taking into account the presence of the layers of gneissic granite mentioned above.

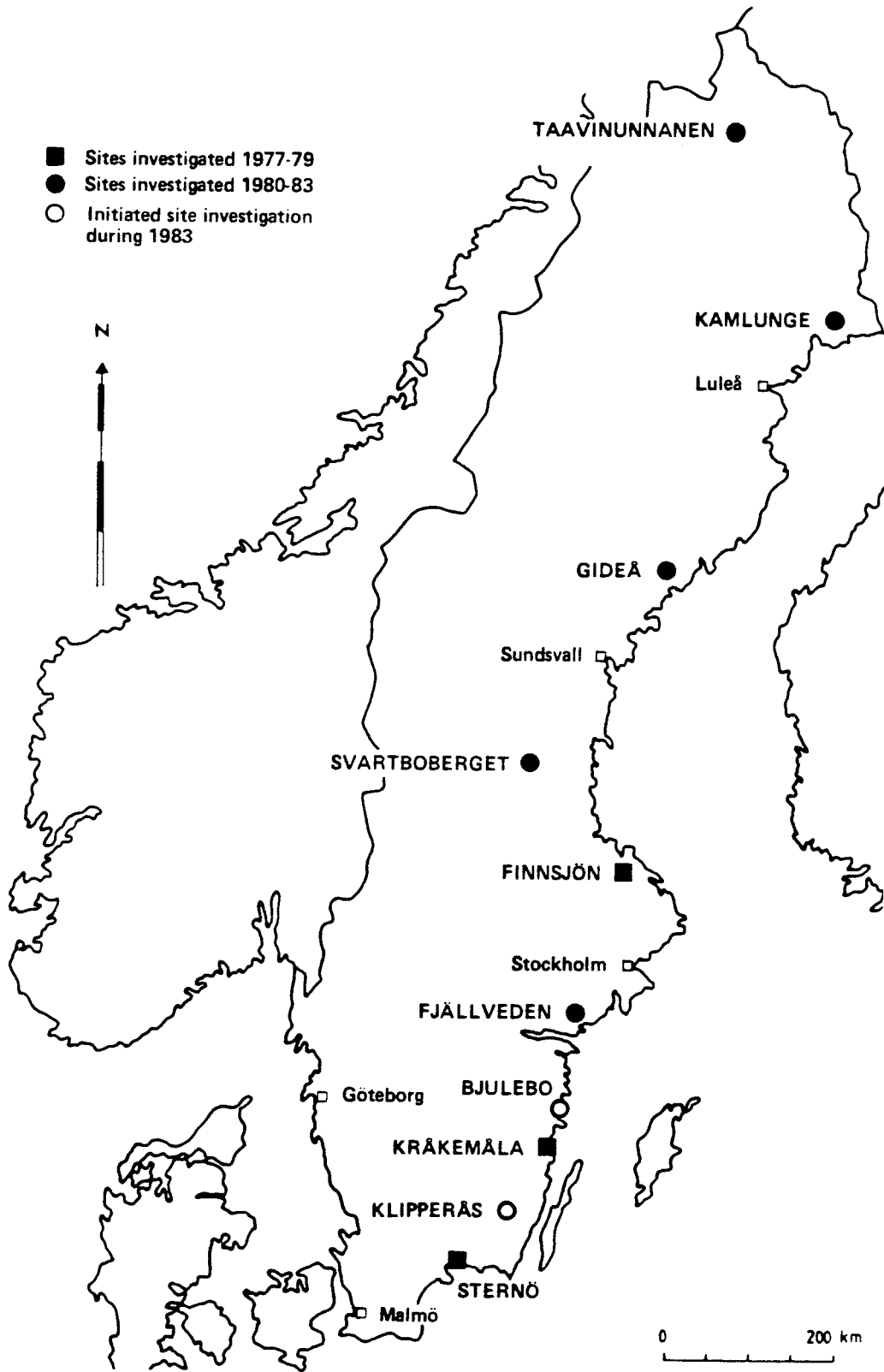


Figure 6-2. Location of study sites.

Table 6-1. Summary of study sites.

Site	Major rock type	Initiation - completion of investigation	Potential for acceptability as repository site
Finnsjön	Granite	1977-79	Low
Kråkemåla	Granite	1977-79	Low
Sternö	Granite	1977-79	High
Fjällveden	Gneiss	1980-83	Reasonable
Gideå	Gneiss	1980-83	High
Kamlunge	Granite	1980-83	High
Svartboberget	Gneiss	1980-83	Low
Tavinunnanen	Gabbro	1981-	?
Bjulebo	Granite	1983-	?
Klipperås	Granite	1983-	?

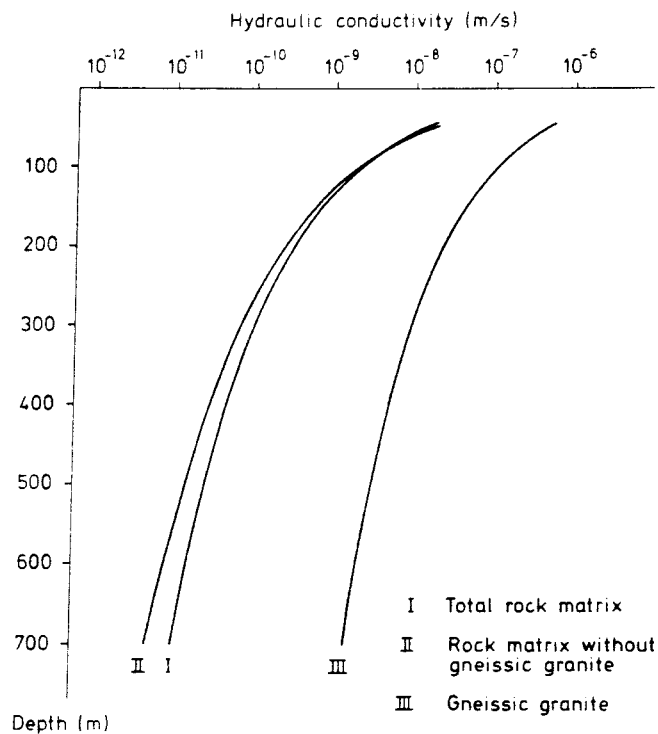


Figure 6-3. Relation between hydraulic conductivity and depth in different rock types at Fjällveden.

6.1.2 Gideå

13 core boreholes have been drilled on the site down to a maximum depth of 700 m. The total length of the core boreholes is about 8 250 m.

The site is characterized by a flat topography (KBS TR 83-53) and the rock mass is dominated by veined gneiss of low hydraulic conductivity. The hydraulic conductivity in the local fracture zones is insignificantly higher than that in the rock mass. As in Fjällveden, granite gneisses of elevated hydraulic conductivity run as layers through the rock mass. In Gideå, however, these layers are oriented horizontally and their hydraulic conductivity differs less from that of the main rock. Diabase dykes in the rock exhibit the same low hydraulic conductivity as the main rock.

The groundwater flow at repository depth in the low conductivity rock has been calculated to be between 0.004 and 0.02 $1/(m^2 \cdot \text{year})$, regardless of whether the influence of the horizontal zones of higher hydraulic conductivity are taken into account or not.

6.1.3 Kamlunge

16 core boreholes have been drilled down to a maximum depth of 670 m on the site. The total length of the core boreholes is about 7 800 m.

The investigated site is shown on Figure 6-4, where the location of a conceived final repository has also been indicated.

The site is situated on a plateau (KBS TR 83-54) with a local relief of about 30 m. The relief to surrounding valleys is about 100 m.

Gneisses and red granite dominate the bedrock. The bedrock also contains amphibolite and granodioritic rock types.

The rock mass possesses low hydraulic conductivity and is intersected by a few narrow, steeply dipping local fracture zones.

A horizontal fracture zone with elevated hydraulic conductivity has been encountered at a depth of about 550 m below the surface. The zone has been penetrated by four core boreholes, where it has been found to have a thickness of between 4 and 14 m.

The groundwater flow at repository depth, about 450 m, has been calculated to be between 0.003 and 0.06 $1/(m^2 \cdot \text{year})$.

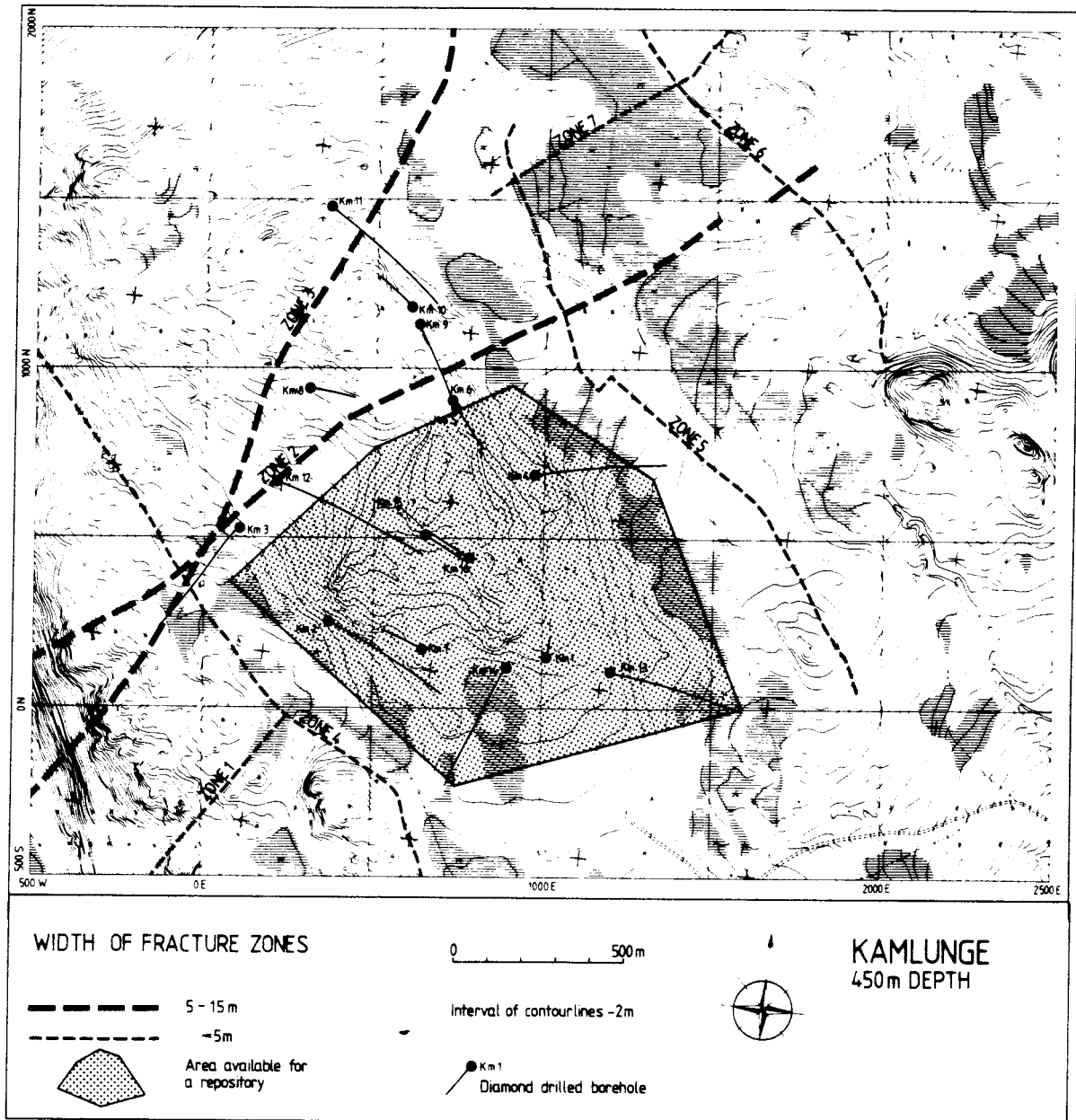


Figure 6-4. The study site at Kamlunge.

6.1.4 Svartboberget

A complete analysis has not been carried out of the suitability of Svartboberget for the location of a final repository. The investigations have indicated local fracture zones spaced at about 300 m and with a dip of about 45° , (KBS TR 83-55). This means that a final repository would have to be divided into several small units, which is not a practical solution.

6.1.5 Tavinunnanen

Only one borehole has been drilled into the gabbro at Tavinunnanen. The depth of the cored borehole is 700 meters. Apart from core logging, also geophysical, hydraulic measurements and water sampling have been performed in the borehole.

The corelogging describes a gabbro intersected by dikes of aplite and granite, in total approximately 8% of the core length. The fracture frequency is normal compared to other investigated sites with a mean fracture frequency of 3.9 fractures/meter.

The magnitude of the hydraulic conductivity at depth is also comparable with other investigated sites. However, the uppermost part of the rock shows lower hydraulic conductivities than the investigated granite and gneissic formations. However, the analysis of the sampled groundwater is unfavourable and gives indications of contamination from near surface waters.

6.1.6 Klipperås

The main field activities were initiated during 1983. The site is characterized by a very flat topography. The widespread thin soil cover makes the surface geological interpretation uncertain. The few existing outcrops are mapped as an homogenous, red mediumgrained granite.

A preparatory geophysical surface investigation was carried out. The area is interpreted as being intersected by only a few fracture zones and some diabase dikes.

An approximately 550 m deep borehole was drilled during 1983. Core logging, geophysical logging, water sampling and hydraulic measurements were carried out.

The fracture frequency obtained from the core logging is relatively high with a mean value of 6.8 fractures/meter. However, the hydraulic conductivity data from 300 meter and to the bottom of the hole were dominated by values below 10^{-11} m/s. The analysis of the obtained water samples shows a favourable composition of the water.

6.1.7 Bjulebo

Surface geological mapping and geophysical measurements were initiated during 1983. However, the field activities were - at least temporarily - cancelled due to a proposal for a new law that prohibits installations related to nuclear power within the actual area along the coast of the Baltic.

6.2 RESEARCH AND DEVELOPMENT

6.2.1 Geology

Neotectonics

A geological documentation of quaternary faults in the northern part of Sweden was completed during 1983 (TR 83-58). The program also included ground- and aerogeophysical investigations (TR 83-57). A great deal of experience on the quaternary faults were gained but further studies on eg. the tectonic explanation of the faults are needed.

Computerized core logging

A microcomputer-based drill core logging system has been developed (TR 83-44). The system is used for registration, storage and presentation of information from the geological core logging, Figure 6-5. Upon completed mapping, all information on the drill hole is transferred to a central computer for storing in the data base. Data are then available for more sophisticated processing, eg. for statistical treatment of fracture frequencies.



Figure 6-5. Equipment for core logging.

6.2.2 Geophysics

Crosshole radar

A crosshole electromagnetic (radar) program has been initiated by SKBF. As of January 1, 1983, the radar program is part of the Stripa Project, see Chapter 10.

Crosshole seismics

Development of seismic crosshole techniques for the detection of fracture zones in crystalline rock has been funded by SKBF. Seismic signals are generated in one borehole by microexplosions while recording is made in another borehole and on the surface. As with the radar program, the development of the crosshole seismics is now part of the Stripa Project, see Chapter 10.

A spin-off from the crosshole seismic program is the detection of tube-waves. The positions of the sources of the tube-waves can easily be correlated to fractures and fracture zones along the borehole wall.

6.2.3 Hydrogeology

Measurement of groundwater flow

A probe for in situ groundwater flow determinations has been developed. The intention of the program is, to some extent, to verify the groundwater flow calculations. A tracer is injected in a sealed off section in a borehole and the groundwater flow through the section may be determined by measuring the dilution of the tracer with time.

The laboratory and initial field test have been very encouraging and further development is in progress.

Measurement of the piezometric pressure

An equipment for an automatic registration of the piezometric (natural) groundwater pressure in 5 different sections in a borehole using one pressure transducer has been developed (TR 83-44, Figure 6-6). The data are transferred via radio and a telephone modem to a main computer.

The equipment may also be used for hydraulic interference tests (pump tests).

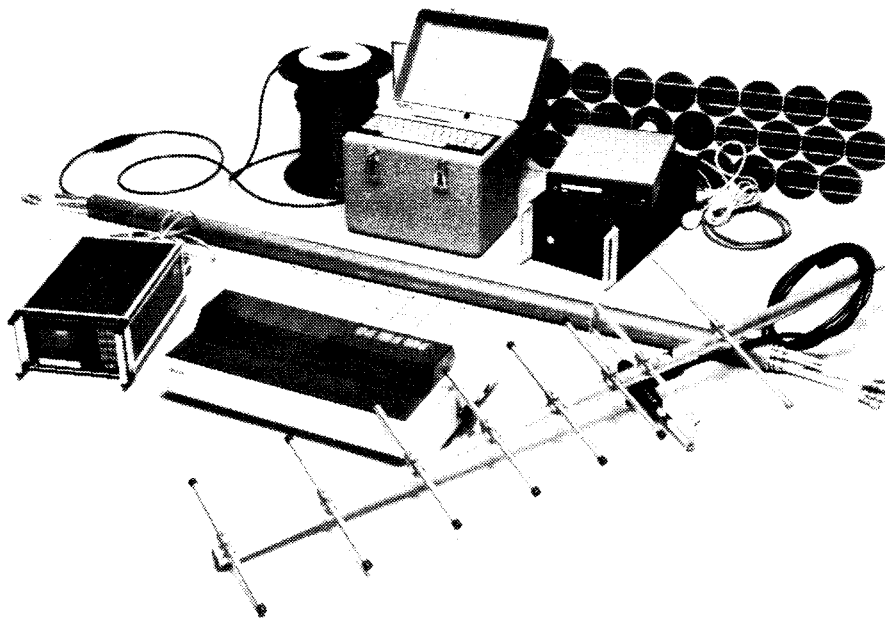


Figure 6-6. Complete piezometric measurement equipment including piezomac, probe, radio, solar cells, tape recorder and printer.

Determination of hydraulic conductivity

The hydraulic conductivity as determined by injection tests in boreholes within the site selection studies is made by using two different systems - a pipe string system and an umbilical hose system. The operation of the pipe string system is much more time consuming and the data handling and presentation is less sophisticated compared to the umbilical hose system. A modification has therefore been initiated in order to improve the pipe string system.

Development of the model for groundwater flow calculations

The model has been further developed to take into account saturated and unsaturated flow which allows for more appropriate description of the present flow problem. Equations for the solution of the coupled flow and stress changes have been implemented and verification is in progress. In addition, work is in progress in order to take separate discrete features in the rock mass, into account.

6.2.4 Hydrogeochemistry

Development of a borehole probe for measurements of Eh, pH and pS^{2-} integrated with a mobile hydrogeochemical field laboratory has been in progress throughout 1983. See 5.2.2.

7 THE BIOSPHERE

The work performed during 1982-1983 has been closely coordinated to the evaluation needs in the KBS-3 report. Thus the biosphere investigations have been concentrated to areas where the geological and hydrological characteristics have been found to be suitable for a possible repository siting.

Two major studies on the natural levels of radioactivity were reported in 1982-83. Three uranium rich springs in northern Sweden has been investigated (KBS TR 82-13). The uranium and radium concentrations during different seasons were measured in spring water, peat and sediments together with data on the springs and their environment, and the concentrations of the major anionic and cationic constituents of the spring water. Investigations of the distribution of uranium, thorium and radium in soil, plant material and drainage water of agricultural lands has also been made (KBS TR 83-73). The chemical and physical characteristics of the soils were determined. Both studies have been part of an effort to build up a basis of information on the behaviour of natural radioactive elements in environments typical for Sweden.

During late 1982 and -83 the emphasis of the biosphere studies was shifted over to site specific data from places under evaluation in the geological site investigation program. In two reports descriptions of the recipient areas (KBS TR 83-11) and the radioactivity levels in surface waters, soils and sediments (KBS TR 82-22) are presented for the Fjällveden, Svartboberget, Gideå and Kamlunge sites. This was made in order to have a realistic data base for the biosphere model used in the safety evaluations.

Other data used in the calculations of nuclide intake are presented in KBS TR 83-28. This report discusses and defines the transfer factors, the concentration and distribution factors, the land use and yield values and the diet and consumption rates used in the BIOPATH model in the KBS-3 evaluations. In KBS TR 82-14 the biologic factors of relevance for the transformation of

intake to absorbed dose are reviewed for isotopes that are of special interest for the long term effects.

For the rest of this decade the biosphere will be mapped in all the areas that are selected for geological site investigations. The mapping will include data on the radioactivity in surface waters and soil and a characterization of the eco-system at hand. Efforts will be made to evaluate the effect on dose calculations of possible changes of the eco-system with time.

8 SAFETY EVALUATIONS

During 1983 the development of a methodology for performance evaluations of final repositories has been strictly goal-oriented toward the intended feasibility proof in the KBS-3 report.

This means that the analysis has been performed on data characterized by high safety margins which in turn give the data base a low internal consistency. Consequently very little effort has been directed to sensitivity and uncertainty assessments in the work for KBS-3.

The selection of scenarios, data and models for the KBS-3 analyses was made with the intention to produce bounding values for the environmental impact of a repository.

The main results are presented in the following reports

Groundwater movements	KBS TR 83-45
Groundwater chemistry	" TR 83-59
Canister corrosion	" TR 83-24
Actinide solubilities	" TR 83-35
Redox front	" TR 83-68
Geosphere transport	" TR 83-48
Biosphere transport and resulting environmental doses	" TR 83-49
Occupational doses	" TR 83-30

To give a perspective on these bounding evaluations, an assessment of the consequences based on nuclide concentrations in natural granitic environments is presented in KBS TR 83-29.

During the coming years efforts will be made to develop a computerized system for performance evaluation and safety assessments, including the use of data of a probabilistic nature and the presentation of the total risk-consequences-spectrum.

9 REPOSITORY FOR REACTOR WASTE – SFR

9.1 GENERAL

In March 1982 SKBF applied for a license to build and operate a central final repository, SFR, for low and medium level wastes from reactor operation. The application was based on a preliminary safety report for a repository located near the Forsmark nuclear power plant. The repository will be situated in crystalline rock under the Baltic sea.

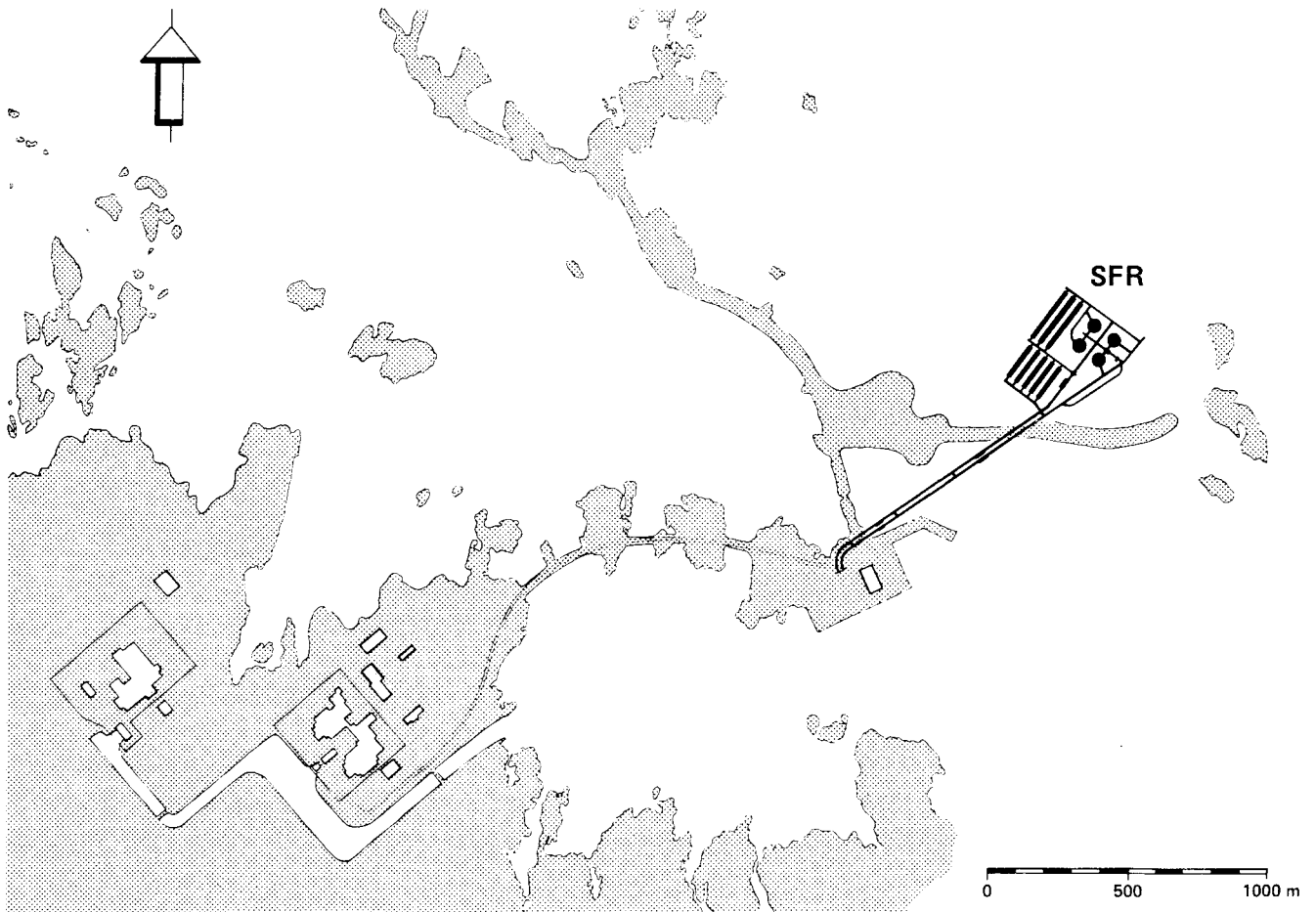


Figure 9-1. Map over the Forsmark site.

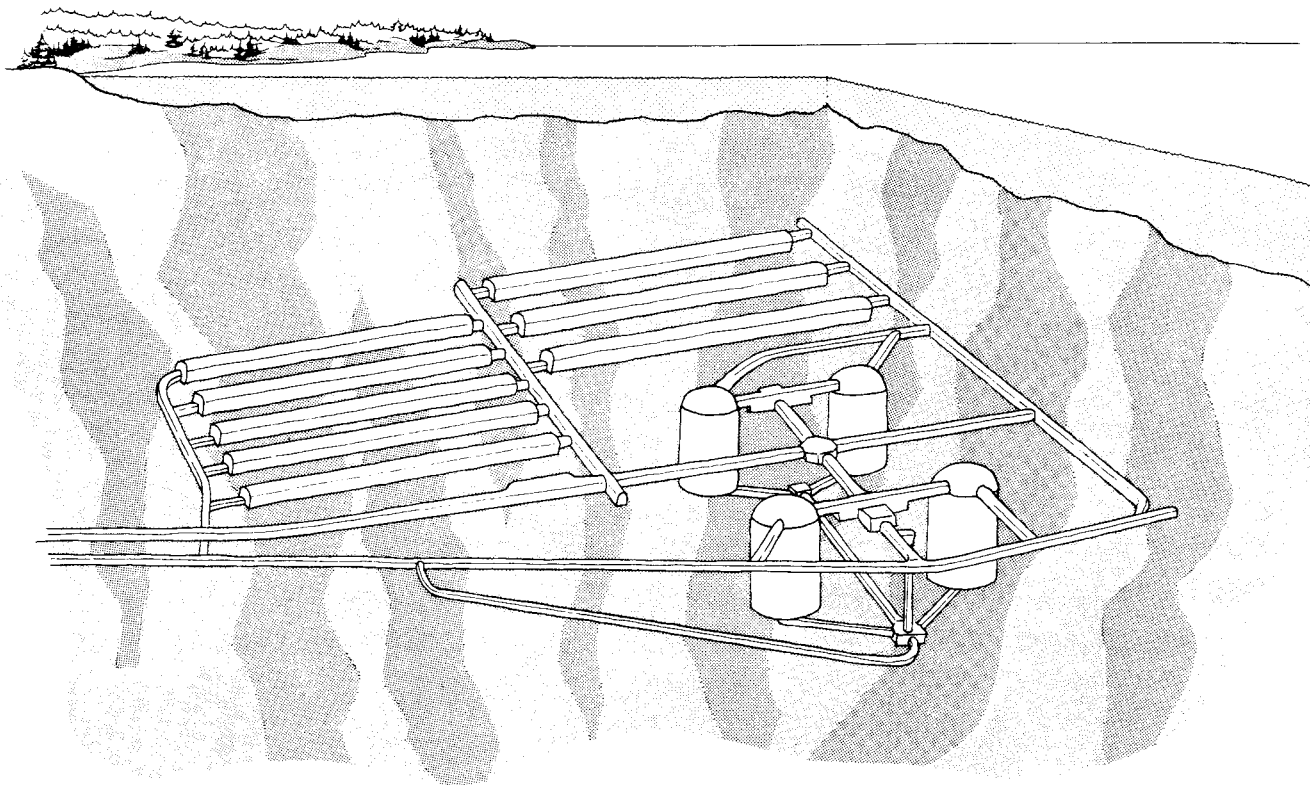


Figure 9-2. General layout of the tunnels and rock caverns.

The safety report was then scrutinized by the Swedish authorities and in June 1983 the Government granted SKBF a construction license for SFR in Forsmark. The license contains some requirements to be fulfilled during the construction period, for instance regarding information to the public about the safety aspects and development and execution of an extensive control and verification program.

The waste to be deposited in SFR mainly consists of short lived low- and medium level waste from the operation of the Swedish nuclear power plants. But also a limited amount of similar radioactive waste from other industries, medical- and research activities will be disposed of in SFR.

9.2 DESIGN AND CONSTRUCTION

The construction work started in July 1983 and the excavation of the two tunnels from the harbour at Forsmark down to the repository level started in October. The caverns will be located 1 km from the harbour and at least 50 m under the sea bottom. The caverns are individually designed for the different types of wastes.

The final layout is based on further geological surveys which were carried out during the autumn of 1983. The result of these surveys confirmed that the rock mass in the area is suitable for the planned caverns.

The storage area can be divided into two types of rock caverns - conventional caverns and silos. The repository will be built in two phases. The first phase which now is under way includes two silos and five caverns. The second phase in the 90's will include another two silos and three caverns. The total storage capacity will then be about 100 000 m³ of waste.

9.3 THE STORAGE AREAS

Most of the activity will be disposed of in the silos. This part is designed as rock caverns, 60 m high and 30 m in diameter. In the caverns 50 m high concrete silos will be constructed. The concrete silos are 25 m in diameter and equipped with internal walls. These walls divide the silo into 2.5 m wide squared shafts. The concrete silos will be surrounded by a buffer of bentonite clay. (Figure 9-3)

The handling of waste packages in the silos are remotely controlled. A special truck brings the waste in a shielding container to an unloading position. The lid of the container is removed by an overhead crane, and the waste packages are unloaded with a handling machine that transports them to one of the concrete silos where they are lowered into the squared shafts. The waste is continuously surrounded by concrete grout.

For the less active waste conventional rock caverns will be used. The design of the rock caverns is dependent on the type and dose rate of the waste packages.

9.4 TIME SCHEDULE AND COSTS

The excavation of the two tunnels had reached 150 m into the rock at the end of 1983. The storage area for the repositories will be reached at the beginning of 1985. When the first caverns are excavated the concrete work gradually will start. The first silo will be constructed in 1986-1987. The repository is planned for operation in 1988.

SKBF has contracted the Swedish State Power Board for the detailed design and the construction of the repository. The total cost

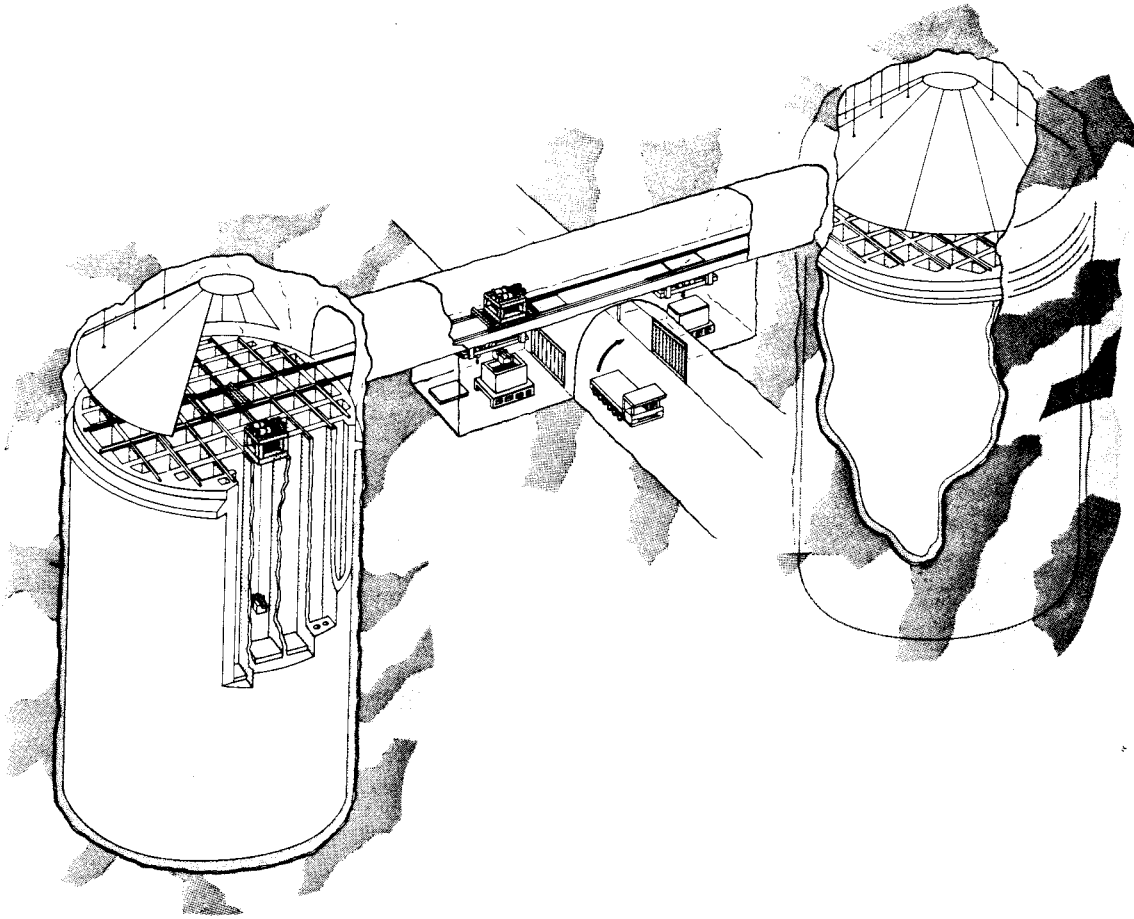


Figure 9-3. Handling of waste packages in the silo repository.

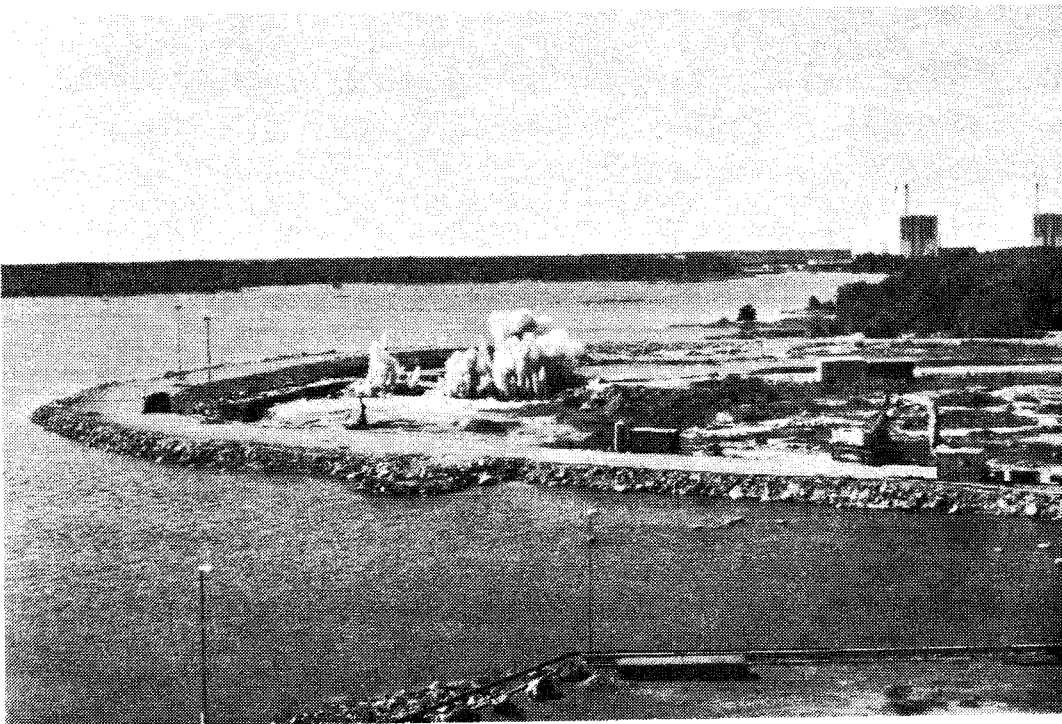


Figure 9-4. In August 1983 the rock-blasting work started for the descent to the tunnels on a small island close to the harbour in Forsmark.

for this work is calculated to be about 900 MSEK (million Swedish kronor) for phase one and two. Costs for operation and sealing (around the year 2025) of the facility have been estimated to be about 300 MSEK. The costs are given in 1982 prices and do not include interest during the construction period.

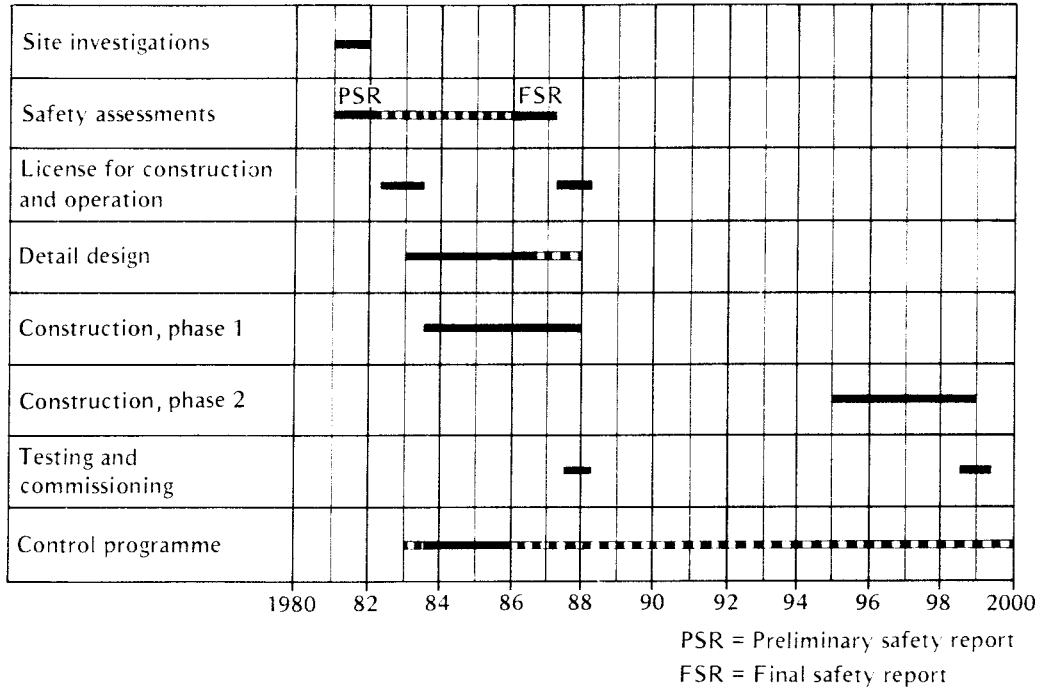


Figure 9-5. Time schedule

10 STRIPA PROJECT

10.1 INTRODUCTION

The Stripa Project is an international cooperative project carried out in an abandoned iron ore mine located in central Sweden. The project concerns research in a realistic environment of different matters connected to disposal of highly radioactive wastes from nuclear power generation, deep underground in crystalline rock. The Stripa mine is only a test station and there is no intention to use or store any radioactive material in the mine.

The project is carried out as an autonomous project under the sponsorship of the OECD Nuclear Energy Agency (NEA) and is managed by the division KBS of SKBF under the direction of representatives from each participating country.

A first phase of the project was initiated in 1980 and a second phase in 1983. The entire project is scheduled to be completed during 1986. Participating countries are Canada, Finland, France, Japan, Sweden, Switzerland, United Kingdom and the United States. Also Spain has declared the intention to join the project.

The experiments are carried out mainly at the 360 meter level in a massive, grey to light red, medium-grained granite.

The progress of the work is presented in a separate annual report. Below a short summary is given.

10.2 RESEARCH PROGRAM 1980–1986

The Stripa Project includes a number of subprojects with different objectives, budgets and time-schedules. Essentially, the research is concentrated to the following areas

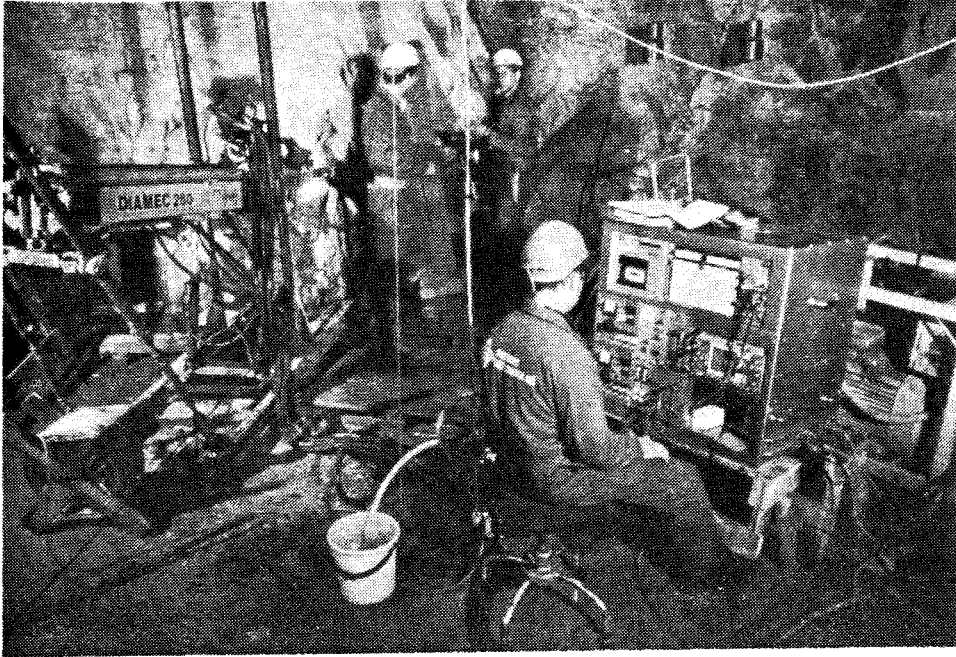


Figure 10-1. Preparatory investigations at the crosshole site at Stripa.

- detection and mapping of fracture zones
- groundwater conditions and nuclide migration
- bentonite clay as backfilling and sealing material

The results obtained are presented in quarterly, internal and technical reports.

An international symposium, where the experiments and obtained results will be presented, is scheduled for early summer 1985.

10.2.1 Detection and mapping of fracture zones

The objective of the investigation is to develop geophysical and hydraulic methods and instruments for the purpose of detecting and mapping of fracture zones. Electromagnetic (radar), seismic and hydraulic (sinusoidal) techniques will be developed and tested.

The investigations are carried out in both single and multiple boreholes, so called crosshole measurements. Different kinds of signals are transmitted through the rock. A disturbance like for instance a fracture zone will affect the transmission. The location and orientation of fracture zones may be determined by per-

forming the tests from a large number of sections on the boreholes.

The investigation started in 1983 and most of the site preparation work was completed by the end of the year. Some preliminary field tests with promising results were also carried out.

Seismic investigations may be carried out at distances up to 1 000 meters or more between the transmitter and the receiver, making the Stripa mine less suitable as test site. In addition to Stripa, the site Gideå, located in the northern part of Sweden, is therefore used as a complementary test site. The geology at the Gideå site has been carefully investigated by SKBF within the program on the selection of a potential repository site for spent nuclear fuel.

10.2.2 Groundwater conditions and nuclide migration

Part of the investigations at Stripa are aimed at developing and testing different methods for determining the hydraulic conductivity of the rock in both vertical and horizontal boreholes. In addition, tests between boreholes, so called interference tests, have been used.

The investigations also aim at characterizing the history of the Stripa groundwater and determine a general program for water sampling and analysis in crystalline rock. Water samples have been taken from a maximum depth of 1 200 meters below the surface and preliminary data indicates that the Stripa groundwater is a mixture of different waters with varying origin and age. Groundwater age determinations is not possible without a detailed knowledge of the hydrogeological and hydrogeochemical characteristics of the site.

Nuclide migration in fissured crystalline rock is a complex phenomenon and has to be investigated thoroughly. An experiment where both sorbing and nonsorbing tracers were injected in a predetermined fracture was initiated in 1980. The distance between the injection point and the sampling points is approximately 5 meters. Core drilling around the injection point gave information on the migration of the sorbing tracers. The experiment is scheduled to be completed in early 1985.

The migration experiment mentioned above is also a preparatory experiment for a large scale migration test, initiated in 1983. An experimental drift, in total 100 meters long, has been carefully blasted, away from the disturbing mining excavations. Non sorbing tracers will be injected at a maximum of nine different locations and at a maximum distance of 50 meters away from the drift. The entire roof and part of the walls are covered with plastic sheets where the inflowing water is collected in separa-

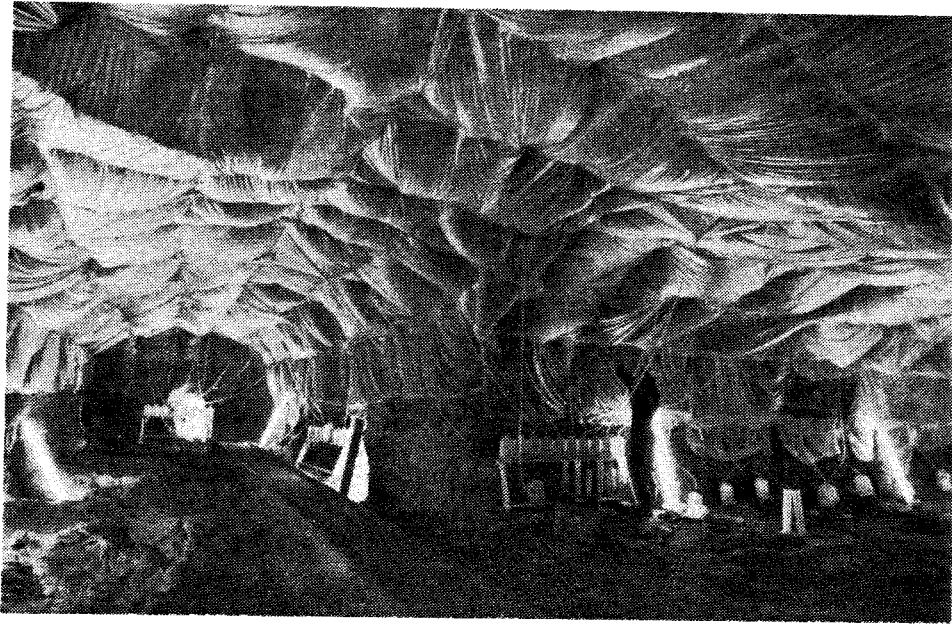


Figure 10-2. Part of the experimental drift used in the large scale migration test and covered with plastic sheets.

BUFFER MASS TEST

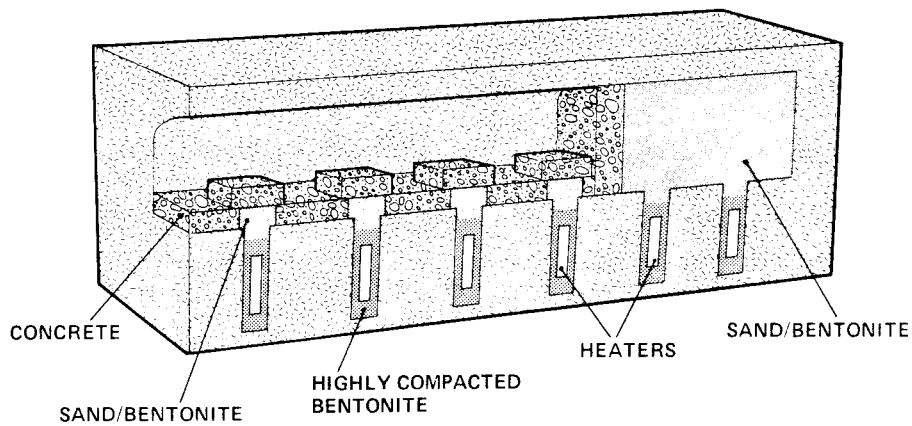


Figure 10-3. Schematic layout of the buffer mass test.

ted sections, see Figure 10-2. Preliminary data indicate that the water flow into the drift is concentrated to three limited areas.

10.2.3 Bentonite clay as backfilling and sealing material

A schematic layout of the buffer mass test, initiated in 1980 and planned to be completed and reported in 1985, is shown in Figure 10-3. Six deposition holes, with a diameter of 0.75 meters and a depth of appr. 3.5 meters, were drilled in a 40 meter long drift. Highly compacted bentonite surrounds the simulated canisters in the boreholes which are overlayed with a mixture of sand and bentonite. The purpose of the test is to verify laboratory data on the bentonite behaviour. Thus, temperatures, water uptake, swelling and water pressures are measured. The heater power was set to 600 W but supplementary tests at elevated power, 1 200 and 1 800 W, are also carried out. The deposition holes are excavated after certain time periods and the bentonite is carefully examined. Figure 10-4 shows the development of total pressure and water pressures in hole no. 2 and the overlying backfill inside the bulkhead.

A number of tests were started in 1983 in order to investigate bentonite clay as sealing materials for boreholes, shafts and tunnels. Figure 10-5 shows highly compacted bentonite inside a perforated copper tube used in the borehole sealing experiments. Figure 10-6 shows the experimental layout of a test where highly compacted bentonite is investigated for the purpose of sealing a highly fractured waterbearing zone crossing a drift. The investigations related to sealing is scheduled to be completed and reported in early 1986.

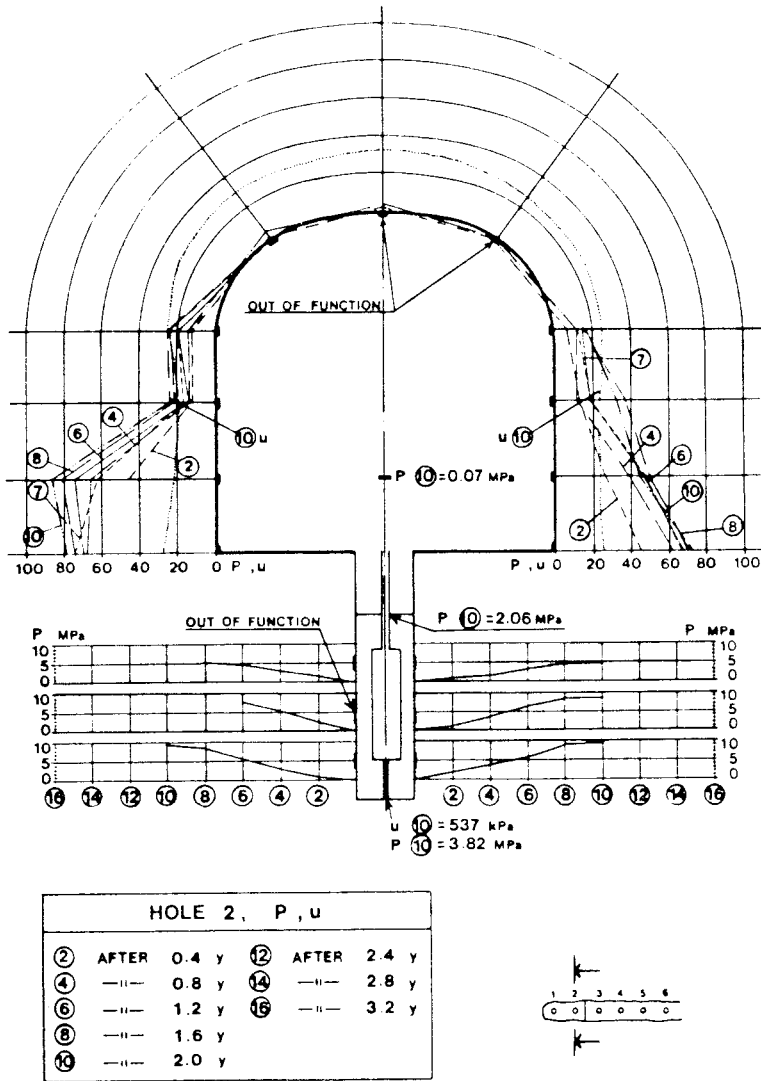


Figure 10-4. Development of total pressures (p) and water pressures (u) in hole no 2 and the overlying backfill.



Figure 10-5. Highly compacted bentonite surrounded by a perforated copper tube used in the overlying backfill.

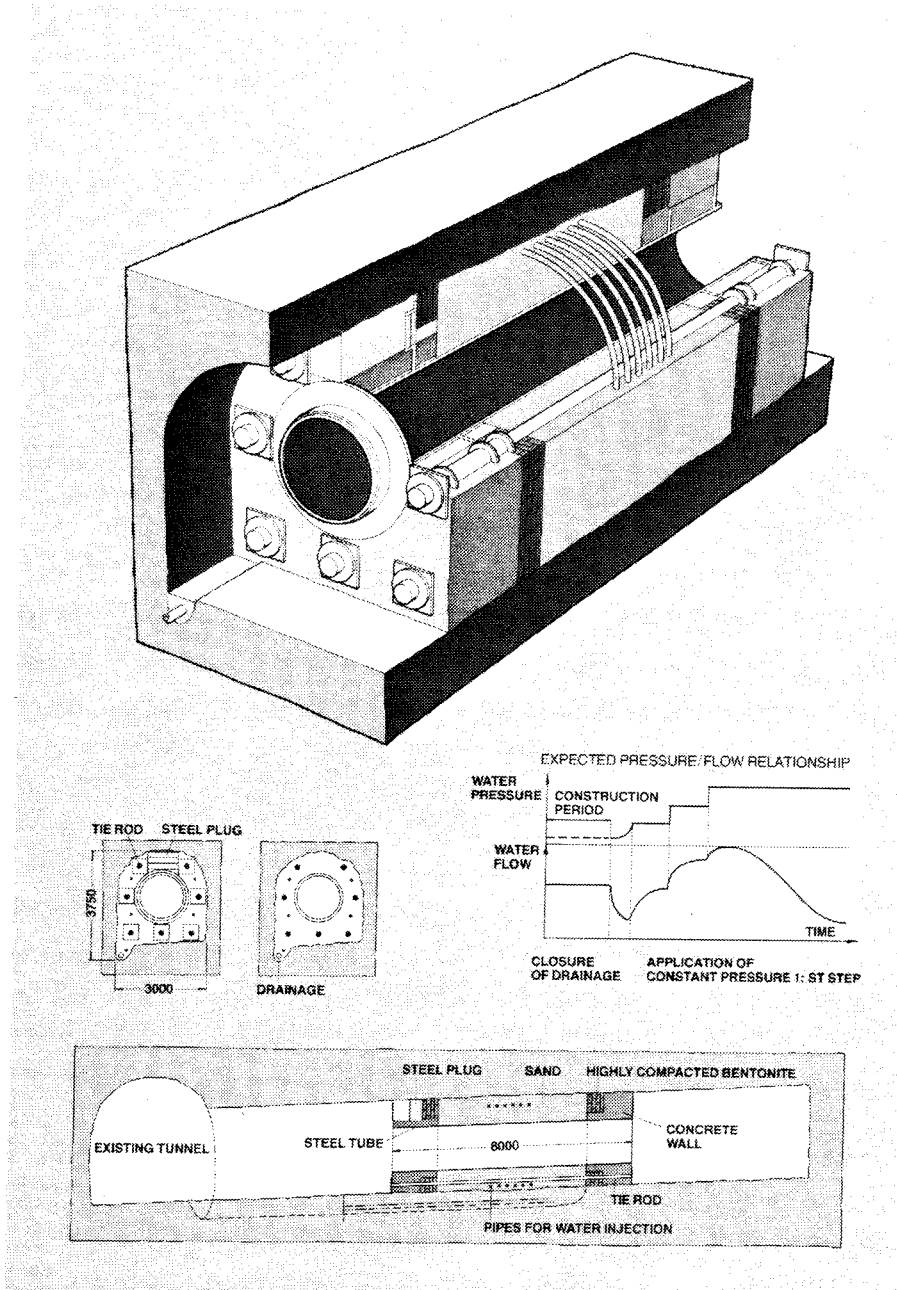


Figure 10-6. Schematic layout of a test. Highly compacted bentonite is tested for sealing a waterbearing fracture zone crossing a drift.

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Swedish Environmental Research Institute
Stockholm, Sweden May 1983

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Thomas Forsberg

University of Luleå

Luleå, Sweden May 31, 1983

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Editor: Lars Werme, SKBF/KBS

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RADIONUCLIDE TRANSPORT IN A SINGLE FISSURE - A LABORATORY STUDY

Trygve E Eriksen
The Royal Institute of Technology, 1983 01 19

SUMMARY

Radionuclide migration has been studied in natural fissures oriented parallel to the axis of granite drill cores. A short pulse of the radionuclides solution was injected at one end of the fissure and the temporal change in radionuclide concentration of the eluate measured. After several hundred fissure volumes water had been pumped through the fissure following the radionuclide pulse the activity distribution on the fissure surfaces was measured. From the retardation of ^{152}Eu , ^{235}Np and ^{237}Pu it is concluded that these radionuclides are transported in the oxidation states Eu(III) , Pu(IV) and Np(V) .

The distribution coefficients K_d calculated from flow and activity distribution data on the basis of geometric surface area/volume ratios are of the same order as published K_d values obtained from batch equilibrium experiments.

THE POSSIBLE EFFECTS OF ALFA AND BETA RADIOLYSIS ON THE MATRIX DISSOLUTION OF SPENT NUCLEAR FUEL

I Grenthe, I Puigdomènech, J Bruno
Royal Institute of Technology, January 1983

SUMMARY

The effects of oxidants on the retainment of actinides in a nuclear repository have been modelled by using an equilibrium procedure. The oxidants are formed as a result of α - and β -radiolysis when spent nuclear fuel is exposed to ground water. From an equilibrium point of view, the strongest reductants in the system (Zr, Pb and Cu) are expected to be oxidized first, leaving the actinoids in the oxidation states they have in the fuel matrix. This is expected to result in a negligible mobilization of the actinoids due to the very low solubility of the MO_2 oxides. However, the formation of protective layers of oxides will most likely decrease the effectiveness of the metallic reducing agents. This will lead to an increased oxidation of the spent fuel which results in an increased actinoid mobilization.

The results of the equilibrium calculations show that the oxidation of the fuel matrix results in the formation of $UO_2(OH)_2(s)$ and to the formation of the soluble complex $UO_2(CO_3)_3^{4-}$. The transport of uranium is limited by the total concentration of carbonate in the aqueous phase.

Neptunium may be quantitatively solubilized as various Np(V) species and transported by ground water from the repository.

Plutonium is retained at the repository site as insoluble PuO_2 . Only very small amounts are transported by ground water.

The mobile actinoids may be reprecipitated when they encounter reducing conditions along the flow path. The conditions for reprecipitation for typical ground water compositions have been modelled by using solubility - pe diagrams.

SMECTITE ALTERATION - PROCEEDINGS OF A COLLOQUIUM AT STATE UNIVERSITY OF NEW YORK AT BUFFALO

Compiled by Duwayne M Anderson
State University of New York at Buffalo, February 15, 1983

SUMMARY

The Colloquium on Smectite Alteration was scheduled over a twoday period with opportunities for individual interaction at several times each day. At the conclusion of the colloquium, participants were invited to summarize their views and provide whatever comments or suggestions that, in their judgment, might prove to be useful to the KBS organization. Some time was allowed for this for participants, who had in the meantime returned to their home institutions. During the month of August informal telephone conversations and correspondence between the colloquium chairman and Drs. Anders Bergström and Roland Pusch of the KBS organization took place. As a result of these exchanges the following questions were identified as of prime interest and importance, answers to which might be included in the conference proceedings then in preparation. These questions can be summarized as follows:

1. What is the prerequisite for and what is the precise mechanism by which smectite clays may be altered to illite?
2. What are likely sources of potassium with respect to the KBS project?
3. Is it likely that the conversion of smectite to illite will be of importance in the 10^5 to the 10^6 year time frame?

During the discussions held at this colloquium a general consensus based upon quite reliable and definitive laboratory and field evidence has emerged. The consensus can be discerned in part or in whole in the individual summaries of the participants. The colloquium has served a highly useful purpose in formulating, and also to a large degree, in validating reliable answers to the three questions posed long before such a consensus could have been expected to emerge in the normal course of events involving the presentation of individual views in scientific journals, presentations at society meetings, etc..

With regard to the first question, after the presentation and subsequent discussion of preliminary views on the subject, there remained little doubt in the minds of the participants that the mechanism for the smectite-illite conversion involves three fundamental steps:

1. The creation of a high lattice charge by the removal of silicon atoms, precipitation of silica in various forms at locations adjacent to the clay lattices, the concomitant replacement of silica in the lattice by aluminum, magnesium, etc., locally available from the dissolution of accessory minerals, or the exchangeable ion complex, etc..
2. Influx of potassium ions derived from local dissolution of potassium-bearing minerals, percolating ground water, etc. to saturate the exchange capacity of the now highly charged smectite intermediate.
3. Irreversible collapse of the high lattice charge, smectite intermediate to the typical nonexpanding illite lattice.

The most critical, rate-limiting step is the production of a highly charged smectite lattice by the progressive substitution of aluminum, etc. for the silica in the smectite lattice referred to in step one. The most important prerequisite to the exchange of lattice silicon for aluminum, etc., is an environmental temperature sufficiently high to allow passage of these ions out of and into the lattice, respectively. There is both laboratory and field evidence to substantiate the conclusion that at temperatures above about 100°C the exchange of silicon and aluminum is relatively rapid and the conversion of smectite to illite, other conditions being favorable, therefore can proceed quite rapidly. It is very likely, however, that the process begins when temperatures exceed about 60°C; but at the lower temperatures, rates of conversion are so slow, as to be almost negligible.

As Dr. Kittrick has pointed out, under laboratory conditions a relatively pure smectite saturated with potassium ions can convert to illite in approximately 10^4 years. Clearly, it is the temperature to which the smectite is subjected that initiates the production of a high lattice charge through the exchange of silica for alumina, etc. and sets the stage for conversion to illite when sufficient potassium is present to bring about lattice collapse. The aluminum required for substitution for silica is sufficiently ubiquitous, available for instance at the corner edges of the smectite sheets, that its availability cannot be regarded as a rate limiting step. Silica removed from the lattice is immediately precipitated and removed from active involvement. Therefore, this process cannot be rate limiting. It is the environmental temperature that is critical in determining whether the smectite-illite conversion is possible and the rate at which it will proceed.

With regard to the source and role of potassium ions in circumstances characteristic of the KBS application, this ion will be present as a result of solution of potassium-bearing feldspar, micas, and other potassium-bearing minerals common in crystalline rocks. The presence of potassium in the circulating ground water at sites selected as possible disposal sites in Sweden has been determined. In the absence of any additional potassium-bearing materials used in the construction of the KBS facility, the main source of potassium then must be the minerals just mentioned and potassium ions would be brought to reaction sites in the compacted smectite buffer material by percolating ground water or by

diffusion. Since irreversible lattice collapse of the smectite to form a potassium-bearing illite requires the presence of a sufficient quantity of potassium ions to nearly saturate the exchangeable ion complex and to penetrate all the interlayer space, the rate at which potassium is supplied by hydraulic permeation or by diffusion is very important. This process too has the possibility of being the rate limiting step.

To summarize and recapitulate the smectite-illite conversion involves the processes of solution, ground water flow and ion diffusion, smectite lattice alteration and mineral reconstitution. The most essential steps are the creation of a high tetrahedral lattice charge brought about by the removal of silica and its replacement by aluminum, or other divalent cations, followed by the uptake and incorporation of potassium ions brought to the appropriate sites by ground water percolation or by ion diffusion. The removal of silica from the tetrahedral sheet and its replacement by aluminum, etc. has been shown to proceed at extremely low rates at temperatures below about 60°C. Above 60°C the rate at which this process can proceed continually increases. At temperatures above 100°C the exchange of silica for the aluminum or certain divalent cations to produce the high lattice charge proceeds quite rapidly and may no longer be the rate limiting step.

The quantity of potassium required to saturate the smectite exchange complex in its state of high lattice charge prior to irreversible lattice collapse is sufficiently great that a plentiful source must be available. In impure smectites the source of potassium may actually be dispersed throughout the clay in the form of silt-sized, potassium-bearing mineral particles such as feldspar, mica, chlorite, etc.. In purified smectites an external source of potassium must be available. In this case potassium ions must be brought to individual reaction sites sequentially, by percolating pore fluids or by the diffusion of potassium ions. At temperatures above 100°C it is very likely that for relatively pure smectite the rate limiting process is the transport of potassium ions by hydraulic flow or ion diffusion from its outer boundary inward.

The rate of probable smectite-illite conversion can be reliably estimated in the KBS application from the above considerations. Assuming that the environmental temperature of the stored nuclear waste and the surrounding buffer material never approaches 100°C.

The most probable rate-limiting step in the smectite-illite conversion process is the rate at which potassium ions are transported into the buffer in quantities sufficient to saturate the exchange complex. Smectite-illite conversion thus, must proceed from an outer boundary inward, assuming that there are no sources of potassium ion within. This emphasizes the need for leak-free nuclear waste containers if potassium ions are a constituent and that the particular smectite clay employed as a buffer material be as nearly free as possible from potassium containing accessory minerals. This condition is necessary to assure that the only source of potassium ion is the surrounding crystalline rock and that the potassium ions will be transported into the buffer material only at concentration levels characteristic of the circulating ground water.

Given these circumstances and conditions it is possible to derive an estimate of the probable stability of the smectite clay. The computations would proceed along the following lines: assume a

smectite buffer material of unit thickness (one meter) placed between walls of massive crystalline rock on either side. Visualize a horizontal column of the smectite through which circulating ground water passes ($1 \text{ cm}^2 \times 1 \text{ m}$). Assume a typical value for the pore volume (a typical value would be in the range of 30 to 40% of the total volume of the element under consideration). To proceed further, it is necessary to assign values to the cation exchange capacity of the buffer material, the concentration of potassium ion in the percolating ground water, and the hydraulic conductivity and density of the compacted bentonite; reasonable values are 70 meq/100 g clay, 10 mg/l potassium ion, 10^{-12} m/sec and 1.9 g/cm^3 , respectively.

The next step is to calculate the total quantity of potassium ion required to eventually bring about collapse of all the smectite lattices in the buffer to the illite configuration. Note that this assumes that the environmental temperature may at some time rise to sufficiently high levels to make the conversion possible. The quantity of potassium ion required is found by multiplying the cation exchange capacity of the smectite by the total quantity of smectite present in the volume element under consideration. Next, it is necessary to calculate the length of time required for this quantity of potassium to pass into the volume element at the flow rates that might be anticipated. This can be estimated from measured values of the hydraulic conductivity of the crystalline rocks typical of the potential KBS sites. If one assumes a unit hydraulic gradient the value of the hydraulic conductivity itself can be employed to roughly estimate the time required to fully saturate the smectite with potassium. This number, representing as it does the most rapid possible rate of conversion given the most unfavorable circumstances, is a highly conservative estimate of the minimum time required for total alteration of the smectite buffer to the nonexpansible illite form.

Calculations performed in this manner yield estimates of millions of years, estimates that greatly exceed the times required by the KBS plan to allow for the natural decline of radiation levels to acceptable levels long before there is any danger of the smectite failing to perform its functions as a buffer material. A sample computation and the resulting estimates are illustrated in Appendix A. Depending upon the degree to which one wishes to consider variations in cation exchange capacity, hydraulic conductivity, porosity, hydraulic gradients, etc., a range of estimates of the probable useful lifetime of the smectite buffer material may be obtained. In all reasonable cases the lifetimes greatly exceed the 10^5 to 10^6 year regarded as necessary in the KBS application.

Reliance on these estimates requires acceptance of the fact, unanimously agreed to by the participants of the colloquium, that in order to bring about the smectite-illite conversion, temperatures approaching or exceeding 100°C are an absolute necessity. Even though the smectite may eventually become fully saturated by potassium ion it cannot spontaneously convert to a nonexpanding illite clay unless the environmental temperatures approach or exceed about 100°C for quite a long period of time. When the exchange complex of a smectite clay is fully saturated with potassium but temperatures do not exceed about 60°C the replacement of silica by aluminum, etc., required to bring about the high lattice charge required for conversion to the illite form, the changes in the properties of the smectite are quite minor. Potassium saturated smectite retains its ability to swell and it is highly impermeable to water. Moreover, it retains the plasti-

city necessary to function effectively as a buffer material. Therefore, the mere saturation of smectite with potassium is not in itself a serious matter. It could become serious only if the temperature of the buffer material were to rise to about 100°C and remain there for an extended period of time.

Let us again review the basic process: If the environmental temperature rises to exceed 100°C, lattice expansion due to the increased energy of atomic vibrations permits the escape of silica from its tetrahedral location in the lattice and its replacement by aluminum or certain divalent cations if they are available. Aluminum ions generally are the most plentiful of these and are always in locations that permit almost instantaneous replacement in the tetrahedral sites vacated by silica atoms with these aluminum ions. The overall rate at which this exchange proceeds is known only in qualitative terms. At temperatures in the range of 60°C and below, this is relatively slow and the formation of the highly charged smectite lattice resulting from this process of the substitution of ions of different charges is sufficiently low to become rate limiting. At temperatures approaching the 100°C range, however, this process is sufficiently rapid that the supply of potassium ions regulates the rate at which individual lattice sheets collapse to the illite configuration and become nonexpanding. The proportion of nonexpanding to expanding layers increases as this process continues until all the smectite lattice layers are converted to the high negative charge form and have taken up quantities of potassium sufficient ultimately to bring about their irreversible collapse to the illite configuration. As this process proceeds, potassium ions in the percolating fluids penetrate ever deeper into the unaltered smectite and the zone of alteration advances inward. By viewing the matter in this way the governing role and the importance of the rate of influx of potassium ion can be fully appreciated. Even though the smectite might be fully converted to the highly charged lattice form by the substitution of aluminum for silicon in the tetrahedral positions, collapse does not occur until a sufficient quantity of potassium ion is present. Without the presence of potassium the high temperature form of the smectite clay would still perform effectively as a buffer material in the KBS application. Looking at the same question from another point of view, in the absence of the high environmental temperatures which are required for the formation of a high lattice charge in the smectite and render it vulnerable to collapse, an influx of potassium ions merely converts the smectite to its potassium form and does not render it ineffective as a buffer. This, then, is the basis upon which it is firmly concluded that under the projected conditions of the KBS storage facility, smectite clays are expected to function effectively as a buffer material over times greatly in excess of the 10⁵ to 10⁶ years required in the KBS application.

In addition to the laboratory data referred to in the discussions of the colloquium participants, field evidence is at hand to confirm the conclusions derived from the forgoing arguments. Altaner et al. (1981) have identified a location in the State of Montana where a pure bentonite (smectite) bed lies between two rock strata that contain potassium-bearing minerals. These rocks were formerly buried and subject to ground water permeation. Hower et al., 1982, have fully characterized this interesting occurrence of bentonite and have studied the extent of alteration of the smectite bed. They have found that illitization is nearly complete at both contacts.

The extent of conversion of smectite to illite was observed to diminish in a roughly exponential manner with distance inward. Independent isotope ratio measurements have established that this bed experienced environmental temperatures for a sustained period of time of 100° to 200°C. The time involved has been reliably estimated to be approximately 3×10^6 years. Alteration in whole or in part has affected roughly half of the smectite in the 2.5 meter thick smectite layer. Clearly the field evidence confirms the conclusion that the smectite-illite conversion, even when the temperatures rise to levels sufficient to create the high lattice charge smectite and K^+ is readily available, is very slow. Clearly it is limited by the rate at which potassium ion can saturate the exchange complex and lead to lattice collapse to the illite configuration. In the case of the Montana bentonite deposit potassium ion must have entered the bed by diffusion since alteration upward from the bottom of the bed almost exactly matches the alteration profile from the top of the bed downward.

Diffusion was considered in an earlier report (Anderson and Fountain 1981) and subsequently by Pusch and his co-workers in a report commenting on this work. Diffusion coefficients ranging from 6×10^{-12} m²/sec to an apparent diffusion coefficient of 1×10^{-20} m²/sec or less are quite possible. Anderson and Fountain (1981) derived very low values of the apparent diffusion coefficient from measurements of calcium ion penetration into various clay matrices. The larger values were derived from field tests in soft illitic clay using calcium hydroxide solution to inject a Ca^{++} tracer (Moum et al., 1968). The large range in values makes it difficult to perform computations that will be convincing to all. However, the conclusions, fully supported by the field evidence cited above, is that potassium permeation by diffusion also is very slow in smectite clay. It is of the same order of magnitude as that which would result from hydraulic permeation.

To summarize, all available evidence is indicative of exceedingly slow permeation by ionic species that might bring about undesirable changes in properties of the smectite buffer material. If environmental temperatures can be kept substantially below 100°C it can confidently be concluded that alteration of the compacted smectite buffer material can be held to negligible levels and that it can be fully relied on to perform its essential functions.

STABILITY OF BENTONITE GELS IN CRYSTALLINE ROCK - PHYSICAL ASPECTS

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University of Luleå, 1983 02 20

SUMMARY

A first estimate of the possible penetration of bentonite from deposition holes with radioactive canisters into rock joints traversing the holes was made a few years ago as part of the KBS research. It showed the importance of the geometrical properties of the joints and of the porewater chemistry with respect to the dispersibility of the expanding clay, but did not yield quantitative measures of the bentonite loss. The present, extended study comprises a derivation of a simple rock model as a basis for calculation of the penetration rate of bentonite and of the groundwater flow rate, which is a determinant of the erodibility of the protruding clay film. This model, which is representative of a gross permeability of about 10^{-8} - 10^{-9} m/s, implies a spectrum of slot-shaped joints with apertures ranging between 0.1 and 0.5 mm. Using this model and an earlier derived physical model for the clay penetration it is concluded that less than 2% of the highly compacted bentonite will be lost into traversing joints in 10^6 years. A closer analysis, in which also Poiseuille retardation and short-term experiments were taken into account, even suggests that the penetration into the considered joints will be less than that. The penetration rate is expected to be 1 decimeter in a few hundred years; probably, the widest joints of the model will be filled with clay to within about 2 decimeters from the deposition holes in the same period of time.

The risk of erosion by flowing groundwater was estimated by comparing clay particle bond strength, evaluated from viscometer tests, and theoretically derived drag forces, the conclusion being that the maximum expected water flow rate in the widest joints of the rock model ($4 \cdot 10^{-4}$ m/s) is not sufficient to disrupt the gel front or the large individual clay flocs that may exist at this front. This hypothesis was tested by conducting pinhole experiments with a somewhat more sophisticated technique than that applied in

conventional earth dam design. Although the expected flow/erosion relationship was not explicitly obtained, these experiments still support the conclusion that erosion will not be a source of bentonite loss.

A "worst case" scenario with a shear zone being developed across deposition holes is finally considered and in addition to this, the conditions in the fracture-rich tunnel floor at the upper end of the deposition holes are also analysed. This study shows that even if the rock is much more fractured than "normal conditions" would imply, the bentonite loss is expected to be very moderate and without substantial effect on the barrier functions of the remaining clay cores in the deposition holes.

STUDIES IN PITTING CORROSION ON ARCHEOLOGICAL BRONZES - COPPER

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University of Stockholm, 1983 01 02

SUMMARY

The study concerns measuring the pitting factor on archaeological bronzes. Special problems have been encountered in this measurement of pitting corrosion, since pitting corrosion can only be studied on relatively well-preserved bronzes, whereas in most cases general corrosion has completely ruined the original surface. The selected material is also skewed due to the fact that the studied material has already undergone one selection, so to speak, in that it has exhibited a corrosion resistance that has enabled it to survive at all. It can be deduced from the many tombs that have been found empty of artifacts or where the only metal artifacts are of gold that only a very small portion of the original archaeological metal material has been preserved through the millenia.

The present material indicates that the time factor has had greater importance for the course of pitting corrosion than e.g. environmental conditions and alloying materials. This can probably be explained by the skewing of the material mentioned above. In other words, due to unstable alloys or a poor preservation environment, the object has not been preserved at all or has only been preserved in the form of fragments that are useless for the purposes of this study.

An interesting find is therefore the low pitting factor in this bronze material, which represents numerous rather impure alloys, i.e. alloys with many trace elements.

INVESTIGATION OF THE STRESS CORROSION CRACKING OF PURE COPPER

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Newcastle upon Tyne, UK, April 1983

GENERAL INTRODUCTION

The Swedish KBS project is concerned with the long term disposal of unprocessed radioactive waste. In order to comply with safety regulations, the waste would be encapsulated and the canisters placed in vertically boreholes, drilled from horizontal tunnels in rock 500 m below the surface. It is proposed that the space around the canisters would be filled with a buffer mass consisting of compacted bentonite. The latter is intended to inhibit penetration of ground water to the container and to act as a chemical buffer (regulating the pH of the water to between 8.5 and 9), as well as providing a soft, pliable bed sufficient to support the canister.

Although a variety of container materials have been considered, the safety requirement has necessitated a conservative approach to the disposal of the early stocks of waste and therefore a thick copper container has been specified. Copper is a noble metal that is not inherently reactive and its general rate of corrosion in water, even in the absence of corrosion films or insoluble corrosion products, is usually low. The thickness of container being specified - 200 mm thick - should be ample to cope with the maximum rate of corrosive attack anticipated. Even allowing for excessive pitting the maximum penetration should be no greater than 60 mm in 10^6 years.

Calculation of rates of penetration on a basis of general corrosion, even with due allowance for any pitting that might be expected, would of course become meaningless if there were any likelihood of stress corrosion cracking in the type of environment surrounding canisters. It was therefore essential to obtain information about the susceptibility of copper to stress corrosion cracking in a suitable range of environments. The only method of obtaining such information from a short-term test is by means of slow strain rate testing in tension.

The normal environment that the canisters would be likely to meet in the rock boreholes would correspond to low salinity ground water as specified by an earlier reference group. ⁽¹⁾ This was

therefore the basis environment chosen for the tests to assess stress corrosion susceptibility of both oxygen-free high conductivity (OFHC) and phosphorus-deoxidised (PDO) copper. The pH of the environment should be in the range 8.0 to 8.5.

Since the maximum temperature expected at the copper-bentonite interface was 80°C, it was decided that the majority of tests at ambient temperature should be backed-up by selected tests at the higher temperature. Although it might be expected that any corrosion of the container was proceeding under de-aerated conditions, because of the slow percolation of water through the ground and the restricted permeation through the bentonite surrounding the canisters, it was decided that susceptibility should be assessed under both aerobic and anaerobic conditions.

There were two main reasons for deviating from the basis ground water in assessing stress corrosion behaviour:

- (1) the possibility of accidental contamination of the boreholes by nitrite, as a result of prior blasting, and
- (2) concentration of certain anions in the ground water would be possible due to evaporation effects from the surface of the container which could attain a temperature of 80°C.

A further variable that was considered was that of potential, since deviations from the normal free corrosion potential in a particular environment could be introduced by changes in surface condition.

Although the majority of slow strain rate tests were conducted at a nominal strain rate in the region of $2.6 \times 10^{-6}/s$, a number of tests involved a faster strain rate of $1.7 \times 10^{-5}/s$, since it is known that stress corrosion effects may depend on the particular strain rate involved. It was confidently felt that the absence of significant effects at these strain rates would be a reliable indication of lack of susceptibility to stress corrosion.

Solutions containing sodium nitrite were expected, from other work on copper⁽²⁾ and 70/30 brass, ⁽³⁾ to promote stress corrosion failure and the work carried out was therefore intended to define a safe upper limit for nitrite contamination.

SORPTION OF RADIONUCLIDES ON GEOLOGIC MEDIA
A LITERATURE SURVEY
1: Fission Products

K Andersson, B Allard
Chalmers University of Technology, 1983 01 31

ABSTRACT

A literature survey on the sorption of fission products on rocks and minerals has been performed. Some sorption data for clays and soils have also been included. The fission products investigated were cobalt, nickel, strontium, cesium, technetium and iodine. Parameters of importance to sorption have been identified and a tabulation of distribution coefficients for groundwater conditions (pH 7-9, low to medium ionic strength) is included in the report.

Some conclusions on the sorptive behaviour of these fission products may be drawn:

For cobalt and nickel the sorption is related to hydrolysis. High sorption is observed at a pH where hydrolysis becomes important and the sorption is high as long as cationic hydrolysis products are formed. When pH is increased neutral or negatively charged hydrolysis products may be formed and the sorption decreases.

For these reactions nuclide concentration CEC (cation exchange capacity) and temperature are of minor importance.

For strontium hydrolysis is of no importance at the normal pH of groundwater, but in groundwater above pH 9 carbonate complexation may occur. For most minerals, the sorption is low, usually with a pronounced pH dependence. Other important parameters are ionic strength and CEC. A nonselective sorption due to electrostatic interactions between negatively charged mineral surfaces and Sr^{2+} seems to occur.

For cesium no hydrolysis may be expected and pH has less importance than for Sr. For most minerals, however, the sorption of Cs is higher than for Sr. Important parameters are nuclide concentration and ionic strength. A selectivity for Cs-sorption is found for some minerals, mostly sheet-silicates.

For technetium sorption is due to a reduction of TcO_4^- to $\text{TcO}_2(\text{s})$ and as anions are poorly sorbed, the sorption is dependent on the redox potential.

Iodine is also anionic and poorly sorbed. Minerals containing ions capable of forming iodides with low solubility (Ag, Mg, Pb etc) are, however, sorbing I^- .

FORMATION AND PROPERTIES OF ACTINIDE COLLOIDS

U Olofsson, B Allard, M Bengtsson, B Torstenfelt, K Andersson
Chalmers University of Technology, 1983 01 30

SUMMARY

The formation and properties of particle fractions in actinide systems (americium(III), plutonium(IV), neptunium(V) and uranium(VI)) have been studied in centrifugation (up to 27000 g), electromigration and diffusion experiments. The influence of physical and chemical parameters such as the storage time (6h - 6 months), temperature (5°C - 70°C), ionic strength of the aqueous phase (0.01 M, 0.1 M, 1 M), nuclide concentration (10^{-7} - 10^{-11} M) and pH (3 - 12) was studied.

Particle fractions (radii larger than 20 nm) were formed both for americium and plutonium in the pH-range of interest (7-9). These particle fractions did not exhibit any apparent enhanced mobility. Reduction of the total concentration in solution due to sorption on the vessel walls had maxima in this pH-range. The removal of both americium and plutonium increased with the storage time.

Essentially no particle fractions were obtained in the neptunium systems.

COMPLEXES OF ACTINIDES WITH NATURALLY OCCURRING ORGANIC SUBSTANCES -
A LITERATURE SURVEY

U Olofsson, B Allard
Chalmers University of Technology, 1983 02 15

SUMMARY

Properties of naturally occurring humic and fulvic acids and their formation of actinide complexes are reviewed. Actinides in all the oxidation states III, IV, V and VI would form complexes with many humic and fulvic acids, comparable in strength to the hydroxide and carbonate complexes. Preliminary experiments have shown, that the presence of predominantly humic acid complexes would significantly reduce the sorption of americium on geologic media. This does not, however, necessarily lead to a potentially enhanced mobility under environmental conditions, since humic and fulvic acids carrying trace metals also would be strongly bound to e.g. clayish material.

RADIOLYSIS IN NATURE:
EVIDENCE FROM THE OKLO NATURAL REACTORS

David B Curtis, Alexander J Gancarz
New Mexico, USA, February 1983

SUMMARY

An examination of the mineralogy of the reactor zones at Oklo shows that they have been significantly altered. The rocks immediately adjacent to these zones are also mineralogically modified with respect to normal uranium bearing rocks. There are also significant chemical changes that correlate with the altered mineralogy. Both changes are interpreted to have been attendant upon the operation of the reactors. The mineralogic changes appear to be the consequence of radiation damage, changes in the bulk chemistry of the system and increased temperatures. Chemical changes were the consequence of convectively circulating fluids that transported elements in and out of the rocks. There were also changes in the electrochemical conditions in the rocks. These changes can most reasonably be attributed to oxidizing and reducing species produced by the radiolysis of water.

We have examined the available data to determine the effects of radiolysis products on the rocks. In addition, we have calculated radiation doses and examined the production of radiolysis products in the fluid phase. Quantification and interpretation of these data lead to the following conclusions: 1) There was a net reduction of iron, probably associated with a net increase in total iron in the rocks of the reactor zones. The reduction of iron was most likely the result of hydrogen produced by the radiolysis of water. 2) Commensurate with the iron reduction, there was an oxidation of uranium and multivalent fission products, resulting in their transport out of the reactor zone. Approximately 10% of the uranium and various proportions of these fission products were removed and redeposited in rocks within a few meters of the reactor zones. 4) The calculated radiation doses from alpha radiation and the inferred hydrogen production suggest an effective radiation yield of 0.06 molecules of hydrogen per 100 eV of energy imparted to the fluid phase. Considering radiation from both alpha and beta sources, the G value for hydrogen production is reduced to 0.01 to 0.002 molecules H₂/100 eV.

KBS Technical Report No 83-11

DESCRIPTION OF RECIPIENT AREAS RELATED TO FINAL STORAGE OF
UNREPROCESSED SPENT NUCLEAR FUEL

Björn Sundblad, Ulla Bergström
Studsvik Energiteknik AB, 1983 02 07

ABSTRACT

A comprehensive study of recipient areas in Fjällveden, Voxnan, Gideå and Kamlungekölen is accomplished. Besides general conditions in Finnsjön and Sternö are discussed.

The recipient areas are defined and their climate, hydrology, bedrock, soil, vegetation, land use and yield from arable land described as well as the yield of fish for the surface water of interest. The potential exposure pathways and model system at the different areas are defined.

Long-term variations of geology, climate, hydrology, land-use, acidification and evolution are described. The possible development of the recipient areas is also discussed.

CALCULATION OF ACTIVITY CONTENT AND RELATED PROPERTIES IN PWR AND BWR
FUEL USING ORIGEN 2

Ove Edlund
Studsvik Energiteknik AB, 1983 03 07

SUMMARY

This report lists (in Table 1) the conditions for calculations of the core inventory for a PWR and BWR. The calculations have been performed using the computer code ORIGEN 2. The amount (grams), the total radioactivity (bequerels), the thermal power (watts), the radioactivity from the α -decay (bequerels), and the neutron emission (neutrons/sec) from the core after the last burnup have been determined.

All the parameters have been calculated as a function of the burnup and the natural decay, the latter over a time period of 0 - 1.0 E07 years. The calculations have been performed for 68 heavy nuclides, 60 daughter nuclides, to the heavy nuclides with atomic numbers under 92, 852 fission products and 7 light nuclides.

The most important results are listed in Tables 4 - 11, which also are the basis of Figures 1 - 8.

Tables 2 - 3 contain the activity (bequerels), per unit mass of nuclear fuel, of the most important nuclides as a function of the decay time for the PWR and BWR conditions considered.

SORPTION AND DIFFUSION STUDIES OF Cs AND I IN CONCRETE

K Andersson, B Torstenfelt, B Allard
Chalmers University of Technology, 1983 01 15

SUMMARY

Concrete has been suggested as a possible encapsulation material for long-term storage of low and medium level radioactive waste. At an underground storage of concrete encapsulated waste, a slow release of radioactive elements into the groundwater by diffusion through the concrete must be considered in the safety analysis. The diffusion may be delayed by sorption reactions on the solid.

A wide range of long-lived radionuclides may be present in the low and medium level radioactive waste. Here, the sorption and diffusion of iodide and cesium in slag cement paste and concrete has been studied. The influence of four different water phases (pore water, groundwater, Baltic Sea water and sea water) as well as the influence of some added species (carbonate, sulphate, and magnesium) has been investigated.

A significant sorption of iodide on cement paste in contact with pore water was observed, indicating that the diffusion may be expected to be retarded in this medium.

For cesium the highest sorption was found for concrete and groundwater. This means that the sorption increases as the concrete is weathered. Low or insignificant sorption was found for the cement paste, indicating that the ballast is responsible for the Cs-sorption.

Carbonization enhances the Cs-sorption by about a factor of 3.

The diffusivity of Cs in concrete and cement paste was determined to between $2 \cdot 10^{-14}$ and $8 \cdot 10^{-14}$ m²/s in pore water (where an insignificant sorption was observed).

The choice of ballast as well as addition of suitable "getters" with high sorption of the long-lived radionuclides might decrease the mass transfer rate through the cement.

THE COMPLEXATION OF Eu(III) BY FULVIC ACID

J A Marinsky

State University of New York at Buffalo, 1983 03 31

SUMMARY

It has been determined that fulvic acid (FA) at concentration levels as low as 10^{-14} molar (~ 20 ppm) on a monomer basis, exists in aqueous media as a well-defined molecular aggregate which exhibits microgel properties. This property of the fulvic acid was not discovered earlier because complication by inadvertent CO_2 dissolution occurred; the consequent disturbance of the acid dissociation equilibrium in these studies was not suspected. It was observed (and reported) earlier, for example, that, during extended examination of potentiometric data obtained in these fulvic acid systems during their stepwise neutralization in batch equilibration studies, pH values slowly drifted with time. Time intervals of 30 to 40 days, at which point pH readings approached a plateau, were considered necessary for equilibration of these systems. As a consequence data analysis was prejudiced by the pH-leveling potential of the additional and unaccounted for reaction between CO_2 , H_2O and OH^- ion when pH measurements, obtained after this prolonged interval, were used to characterize the potentiometric behavior of the FA.

The unambiguous deduction that FA exists as a separate phase (microgel) when dispersed in aqueous media is based on the following observations: (1) In studying the distribution of Eu^{154} between 8% crosslinked Dowex 50 resin in the NA^+ -ion form and fulvic acid at several different NaCl concentration levels and over an extended pH range from 4 to as high as 10 it has been shown that only one FA-complexed species of Eu^{+3} , $[\text{Eu}(\text{COOR})]_n^{++}$, is formed, the magnitude of its apparent constant of formation, $\beta_{\text{Eu}(\text{COOR})^{++}}$, being roughly proportional to the third power of the NaCl concentration employed in the study (a direct proportionality is blurred by expansion or contraction of the microgel as experimental parameters change) and (2) although the apparent pK of the fulvic acid, because of significant aberration of the pH data by CO_2

sorption has been quantitatively measured in only a few instances. An inverse relationship between apparent pK and ionic strength has been conclusively demonstrated.

Observations (1) and (2) can only correlate with the fact that the concentrations of free mobile ion counterions in solution are a reflection of the equilibrium between H^+ , $COOR^-$, Eu^{+++} , $HCOOR$ and $Eu(COOR)^{++}$ in the microgel defined by the FA aggregate which controls the concentration ratio of H^+ , Eu^{+++} and Na^+ in the aqueous phase. Thus, if Na^+ , the potential determining ion, is reduced or raised in solution by a factor of 10 in parallel sets of experiments, the quantity of H^+ and Eu^{+++} in solution is reduced or raised, about a factor of $\sim 10^1$ and $\sim 10^3$, respectively, so that the Donnan equilibria between these diffusible components in the microgel and aqueous phases ($EuCl_3$ and $NaCl$, and HCl and $NaCl$) can be fulfilled; the exact proportionality is effected by microgel flexibility.

With the unique description of the FA, Eu(III), NaCl system provided by this useful gel model it has been possible to compare the complexation potential of $CO_3^{=}$ and FA as a function of ionic strength, the particular FA source, pH, and total $CO_3^{=}$. It is quite apparent that some FA sources can be expected to complex Eu(III) so selectively in the presence of $CO_3^{=}$ ion that even as little as 2 ppm of FA can preferentially bind to the Eu(III) in typical ground water systems.

DIFFUSION MEASUREMENTS IN CRYSTALLINE ROCKS

Kristina Skagius, Ivars Neretnieks
Royal Institute of Technology, 1983 03 11

SUMMARY

The diffusion of non-sorbing species in different rock materials and fissure coating materials have been studied on a laboratory scale. The non-sorbing species were iodide, Uranine and Cr-EDTA. The results show that the effective diffusivity for iodide in granites with fissure coating material is of the same magnitude or higher as the effective diffusivity for iodide in granites without fissure coating material. The results also show that it is difficult to give just one value of the diffusivity in a rock material from a certain area because of the variations in the rock material. The effective diffusivity for iodide in rock materials without fissure coating material was determined to be from $1 \cdot 10^{-14} \text{ m}^2/\text{s}$ to about $7 \cdot 10^{-13} \text{ m}^2/\text{s}$.

To simulate the overpressure that exists in the bedrock at large depths, diffusion experiments with iodide in rock materials under pressure have been started. No results from these experiments can yet be presented.

Electrical resistivity measurements in salt-water-saturated rock cores which are under pressure have been performed. The resistivity is measured in the saturated core at various pressures, and in the salt solution with which the core has been saturated. The ratio between these two resistivities has a direct relation to the ratio of the effective diffusivity for a component in the rock material and the diffusivity in free water for the same component.

Results from two measurements are presented and they show that the effective diffusivity is reduced by about 60% at 230-240 bar from the value at atmospheric pressure. More experiments must be made before any conclusions can be drawn.

STABILITY OF DEEP-SITED SMECTITE MINERALS IN CRYSTALLINE ROCK -
CHEMICAL ASPECTS

Roland Pusch
University of Luleå, 1983 03 30

SUMMARY

A recent survey of possible conditions and mechanisms of smectite alteration, with special respect to the Swedish concept of radioactive waste disposal, has shown that the charge change by replacement of tetrahedral Si by Al is the key mechanism of the only practically important alteration, namely that of smectite/illite conversion. If K is available in sufficient quantities it will be fixed and permanent conversion to the unwanted illite-type minerals is a fact; if not, the smectite will be beidellite-like with practically unchanged physico/mechanical properties. Heating to more than about 100°C is thought to be the cause of the charge change.

One other process may be critical and that is cementation of various substances. A possible cementation mechanism, i.e. that of quartz precipitation, is very probably associated with the smectite/illite conversion.

Practical examples of smectite alteration and survival under reasonably well documented geological conditions with respect to temperature and pressure are available, one being that of the Kinnekulle bentonites, another one, although less well known, being the smectitic clay beds in the Höganäs depression. Rather comprehensive core sampling was made at both sites and elemental and mineral analyses were conducted as well as microstructural studies. They support the hypothesis that practically important charge change through Si/Al replacement requires a temperature of more than 100°C, and that such replacement does not yield permanent lattice collapse unless K is available in sufficient quantities. The Höganäs case also serves as an example of drastic loss in plasticity and swelling potential by cementation of other precipitates than quartz, namely iron compounds.

ANALYSIS OF GROUNDWATER FROM DEEP BOREHOLES IN GIDEÅ

Sif Laurent
Swedish Environmental Research Institute, 1983 03 09

SUMMARY

Groundwater from two boreholes in granitic rock at an investigation site in Gideå has been sampled and analysed. This is part of a larger program of geological, geophysical and hydrogeological investigations aimed at finding a suitable site for a high level radioactive waste repository.

Five water-bearing levels in each borehole down to the deepest at about 500 m in the first and to about 600 m in the second borehole were selected. Prior to sampling, the water-bearing level is isolated between packer sleeves. The water is then pumped to the surface where sensitive parameters such as redox potential, pH, sulphide and oxygen content are measured electrochemically on the flowing water in a system isolated from the air. Water, filter and gas samples are sent to several laboratories for further analysis.

The present report is a presentation of the results of the groundwater analyses. The reliability of the results is discussed but there is no evaluation in relation to geology and hydrogeology. This report presents the basic results from the groundwater analyses to be further evaluated by experts in different fields.

MIGRATION EXPERIMENTS IN STUDSVIK

O Landström; Studsvik Energiteknik AB
C-E Klockars, O Persson, E-L Tullborg, S-Å Larson; Swedish Geological
K Andersson, B Allard, B Torstenfelt; Chalmers University of Technology
1983 01 31

ABSTRACT

Field experiments (two-well pulse method) on the migration of Sr and Cs in a gneiss at Studsvik on the Swedish east coast as well as supporting laboratory measurements on sorption/migration have been performed.

Interconnected fractures at about 100 m depth between boreholes were located and characterized. In the boreholes measurements of resistivity, spontaneous potential, pH, Eh, curvature, temperature, gamma radiation, and radon have been performed. The fracture zones were localized, using TV-logging, and the mineralogy of the water bearing fractures in a drill core from the area determined. The main fissure filling minerals were chlorite, calcite and smectite.

The groundwater composition was analyzed and it was found that the water in some cases is oversaturated with respect to calcium carbonate. Temperatures up to 12°C of the pumped water have been observed, corresponding to water originating from 200-400 m depth.

In laboratory batch measurements it was found that the distribution coefficient for Studsvik gneiss and possible fissure filling minerals was 0.01-0.4 m³/kg. The sorption isotherm for the gneiss was almost linear, a Freundlich isotherm with the exponent 0.89 fits well to the data.

In two field experiments, performed between different boreholes, retention factors for Sr of 17 and 30, respectively, were obtained. In laboratory column experiments, performed on crushed borehole material, the retention factor was 30-35, corresponding to a distribution coefficient of 0.006-0.008 m³/kg.

For Cs, injected simultaneously with the Sr, the retention factor is >30. (No breakthrough after 5000 h.)

In both the field experiments a minor amount of the Sr was migrating almost without retention (retention factor <2). It does not seem probable that there should be an extremely short flow path, transporting minor amounts of the water in both the experiments, explaining the fast migration, as different boreholes were used. A more probable explanation is that some of the Sr is sorbed on particles or complexed by organics in the water and transported in one of these forms. A precipitation of Fe is for instance observed when aereating the groundwater, indicating the possibility of Fe-colloid formation. This effect was not observed for Cs, indicating that the process causing the fast transport of Sr is selective to Sr.

ANALYSIS OF GROUNDWATER FROM DEEP BOREHOLES IN FJÄLLVEDEN

Sif Laurent
Swedish Environmental Research Institute, 1983 03 29

SUMMARY

Groundwater from three boreholes at an investigation site in Fjällveden has been sampled and analyzed. This is part of a larger programme of geological, geophysical and hydrogeological investigations aimed at finding a suitable site for a high level radioactive waste repository.

Four water-bearing levels in each of the boreholes FJ2 and FJ4, and two levels in borehole FJ8, have been sampled. The sampling depths range from 106 down to 562 m. Prior to sampling, the water-bearing section is isolated between packer sleeves. The water is then pumped to the surface where sensitive parameters such as redox potential, pH, sulphide and oxygen content are measured electrochemically on the flowing water in a system isolated from the air. Water, filter and gas samples are sent to several laboratories for further analysis.

The present report is a presentation of the results of the groundwater analyses. The reliability of the results is discussed but there is no evaluation in relation to geology and hydrogeology. This report presents the basic results from the groundwater analyses to be further evaluated by experts in different fields.

ENCAPSULATION AND HANDLING OF SPENT NUCLEAR FUEL FOR FINAL DISPOSAL

B Lönnerberg, ASEA-ATOM
H Larker, ASEA
L Ageskog, VBB - SWECO
May, 1983

SUMMARY

The handling and encapsulation of spent fuel for final disposal is dealt with in this report.

The handling and embedding of those metal parts which arrive to the encapsulation station with the fuel is also described.

For the encapsulation of fuel two alternatives are presented, both with copper canisters but with filling of lead and copper powder respectively. The sealing method in the first case is electron beam welding, in the second case hot isostatic pressing. This has given the headline of the two chapters describing the methods: Welded copper canister and Pressed copper canister.

Chapter 1, Welded copper canister, presents the handling of the fuel when it arrives to the encapsulation station, where it is first placed in a buffer pool. From this pool the fuel is transferred to the encapsulation process and thereby separated from fuel boxes and boron glass rod bundles, which are transported together with the fuel.

The encapsulation process comprises charging into a copper canister, filling with molten lead, electron beam welding of the lid and final inspection. The transport to and handling in the final repository are described up to the deposition and sealing in the deposition hole. Handling of fuel residues is treated in one of the sections.

In chapter 2, Pressed copper canister, only those parts of the handling, which differ from chapter 1 are described. The hot isostatic pressing process is given in the first sections.

The handling includes drying, charging into the canister, filling with copper powder, seal lid application and hot isostatic pressing before the final inspection and deposition.

In the third chapter, BWR boxes in concrete moulds, the handling of the metal parts, separated from the fuel, are dealt with. After being lifted from the buffer pool they are inserted in a concrete mould, the mould is filled with concrete, covered with a lid and after hardening transferred to its own repository. The deposition in this repository is described.

AN ANALYSIS OF THE CONDITIONS OF GAS MIGRATION FROM A LOW-LEVEL RADIOACTIVE WASTE REPOSITORY

C Braester, Israel Institute of Technology, Haifa, Israel
R Thunvik, Royal Institute of Technology
November 1982

SUMMARY

The present report deals with the conditions under which gas will migrate from a radioactive low-level waste repository located in a hard rock formation below the sea bottom. Chemical reactions resulting in gas (hydrogen) production will take place in the stored waste. Unless the gas produced can escape from the rock cavern an over-pressure that may bring about contamination of the groundwater regime may arise in the cavern. Therefore, it is of significant interest to investigate the transport properties of the surrounding rock formation and to study how the transport capability compares with the rate of the gas production. The purpose of the present study is to provide a rough estimate of the capability of the rock formation in consideration to convey the gas produced in the repository to the surface. The gas produced in the water filled repository will be subject to very fast gravitational segregation, and as a result a gas cushion will be created at the roof of the cavern. Three particular stages may be distinguished in the present flow problem, these are: (i) the accumulation of the gas at the roof of the cavern, (ii) the escape of the gas into the fractures, once the critical thickness of the gas cushion has been reached, and (iii) the flow of gas, once the water in the fractures has been displaced. A first part of the study is devoted to analysing the transport properties of a single fracture. The governing flow equations and expressions for the flowrates of the gas and break-through times of the gas front to reach the upper boundary (sea bottom) of the rock formation are derived. The physical properties of a single fracture are used for the modelling of the fractured rock mass in consideration. A second part of the study is devoted to a number of numerical examples worked out to study the conditions of the gas flow for the projected low-level radioactive waste repository. The following parameters are studied: (i) the critical thickness of the gas cushion, (ii) break-through times of the gas, and (iii) flowrates of the gas.

CALCULATED TEMPERATURE FIELD IN AND AROUND A REPOSITORY FOR SPENT NUCLEAR FUEL

Taivo Tarandi
VBB, April 1983

SUMMARY

Temperature distribution in and around the final storage has been calculated for BWR-fuel.

The results are also applicable to PWR-fuel if the amount of fuel is adjusted so that the power per canister is the same.

The calculations are made with the conservative assumption of the coefficient of thermal conductivity of 0.75 W/(m °C) in the bentonite and 3.0 W/(m °C) in the rock.

The amount of BWR fuel is about 1.4 ton per canister. The canisters are deposited 40 years after withdrawal from the reactor.

A number of different layouts in single and two-level storages have been studied.

Finally, a two-level storage has been chosen as a basis for further project work.

The maximum temperature increase of 59.2°C at the surface of the canister is reached about 30 years after the time of deposition. However, in this two-level storage there will be also a second temperature peak of 58.7°C about 600 years after the deposition. The highest temperature increase in the rock, 56.8°C, occurs about 600 years after the deposition.

At the same time as the temperature continues to sink, there is a levelling out of the local temperature differences in the storage. These differences are negligible after about 1000 years.

After 100 000 years the temperature in the storage is only a few degrees centigrade above the initial rock temperature.

The heat from the storage reaches the ground surface about 200 years after the deposition. The maximum heat flow, 0.28 W/m², occurs about 2000 years after deposition and is considered insignificant compared for example with solar energy flow of about 100 W/m².

PREPARATION OF TITANATES AND ZEOLITES AND THEIR USES IN RADIOACTIVE
WASTE MANAGEMENT, PARTICULARLY IN THE TREATMENT OF SPENT RESINS

A Hultgren (editor), C Airola, Studsvik Energiteknik AB
S Forsberg, Royal Institute of Technology
L Fälth, University of Lund
May 1983

SUMMARY

Work on the preparation of titanates and zeolites was started and their possible uses in the management of radioactive wastes proposed in the first years of the 1970's by the Department of Nuclear Chemistry at the Royal Institute of Technology in Stockholm and the Chemical Center of the University of Lund, respectively. The major part of these efforts was sponsored by the National Council for Radioactive Waste (Prav), while the concluding phase including an over-all system design study adapted to the Swedish nuclear power programme for an economic evaluation was sponsored by the KBS Division of the Swedish Nuclear Fuel Supply Co.

The preparation work resulted in processes ready for industrial scale production of sorbents of qualities adequate for applications in radioactive waste treatment. The essential effort was devoted to the treatment of spent resins from nuclear power plants by transfer of their radioactive contents to titanates and zeolites, which can then be sintered to stable ceramic bodies (the PILO process). The economic evaluation indicated a significant economic incentive for the introduction of the PILO process if an incineration step is included for all types of spent resins.

The essential efforts and results from this programme are summarized in this report.

CORROSION RESISTANCE OF A COPPER CANISTER FOR SPENT NUCLEAR FUEL

The Swedish Corrosion Research Institute and its reference group
April 1983

Documents included in the report:

- Appendix 1 Composition of the groundwater
Ingmar Grenthe
- Appendix 2 Oxidation of copper by sulphate
R Hallberg och I Grenthe
- Appendix 3 Thermodynamic estimates of the solubility of copper in
contact with groundwaters of different composition
Tom Wallin
- Appendix 4 Approximate calculation of what happens with the
oxygen that is entrapped in the repository at the time
of its closure
Ivars Neretnieks
- Appendix 5 Approximate calculation of corrosion of copper
canister caused by sulphate and sulphide attack
Ivars Neretnieks
- Appendix 6 Corrosion of copper from lightning conductor plates
R Hallberg, A-G Engvall, T Wadsten
- Appendix 7 Investigation of corrosion damage on lightning
conductor plates
Göran Sund, Bo Rendahl
- Appendix 8 Microscopic investigation of the structure of copper
specimens
Sten Modin, Helfrid Modin
- Appendix 9 The corrosion resistance of copper canisters for spent
nuclear fuel - follow-up
Supplementary statement
Gösta Wranglén

FEASIBILITY STUDY OF ELECTRON BEAM WELDING OF SPENT NUCLEAR FUEL CANISTERS

A Sanderson, TF Szluha, JL Turner, RH Leggatt
The Welding Institute Cambridge, UK, April 1983

SUMMARY

A thick walled copper container is presently the prime Swedish alternative for encapsulation of spent nuclear fuel. In order to demonstrate the feasibility of encapsulating high-level nuclear waste in copper containers, a study of electron beam welding of thick copper has been performed. Two copper qualities have been investigated, oxygen free high conductivity (OFHC) copper and phosphorous desoxydized high conductivity copper (PDO). The findings in this study are summarized below.

In 100 mm thick copper penetration can be achieved at power level of about 75 kW (typically 150 kV x 500 mA) at welding speed of 100 mm/min. The welds in OFHC copper made under these conditions are free from major defects during constant welding conditions. The welds in PDO copper show a microporosity level considerably higher than those in OFHC copper, but no major defects are produced in the welds in PDO copper.

In the ending of the weld (ie the fade out) it is still not possible to completely eliminate root and cold-shut defects.

A semi-full-scale lid weld has been performed successfully.

Automatic ultrasonic C-scan has been shown to be useful in detecting and displaying defects, but some problems still remain with defect sizing. The different specimens of OFHC copper had different attenuation of the ultrasonic signal, forged copper showing a far lower attenuation than hot extruded copper, indicating that attention must be paid in choosing copper that allows accurate ultrasonic testing.

Residual stresses in the welded zone has been measured and are found to lie in the range -32N/mm^2 to $+36\text{N/mm}^2$. The peak stress was less than half the assumed value of the proof stress of the fused metal.

THE KBS UO₂ LEACHING PROGRAM
Summary Report 1983 02 01

Roy Forsyth
Studsvik Energiteknik AB, February 1983

ABSTRACT

The results so far obtained in the current KBS program on the leaching/corrosion of spent UO₂ fuel are presented and discussed.

Fuel with a burnup of about 40 MWd/kgU from a fuel rod from the Oskarshamn I BWR is used as source material.

Experiments have been performed mainly under oxidizing conditions, results being obtained which are in broad agreement with previously published work. An interesting feature of the experiments is the determination of the colloidal fraction of the material present in the aqueous phase after contact with the leachant. Such information, together with results of leaching at lower pH values, leads to the hypothesis that the concentrations of uranium and the activities in the aqueous phase are limited by solubility/adsorption effects.

A number of experiments have also been performed where preliminary attempts to impose the more realistic reducing conditions on the system have been made. Here, a small but significant reduction in the apparent leach rates has been achieved.

RADIATION EFFECTS ON THE CHEMICAL ENVIRONMENT IN A RADIOACTIVE WASTE REPOSITORY

Trygve Eriksen, Royal Institute of Technology
Arvid Jacobsson, University of Luleå
1983 07 01

SUMMARY

The radiolytic hydrogen production in compacted bentonite have been measured at two dose rates 6.1 and $0.014 \text{ rad}\cdot\text{sec}^{-1}$. The hydrogen production depends on the Fe^{2+} and HCO_3^- concentration in the pore water and on the dose rate. The hydrogen production at $6.1 \text{ rad}\cdot\text{sec}^{-1}$ is in agreement with the hydrogen production calculated assuming homogeneous kinetics and Fe^{2+} and HCO_3^- concentrations of $2.2\cdot 10^{-7}$ and $6.5\cdot 10^{-5} \text{ mol}\cdot\text{dm}^{-3}$ respectively. The amount of divalent iron in the bentonite clay accessible for scavenging of oxidative radicals was found by Mössbauer spectroscopy to be at least 0.4%.

AN ANALYSIS OF SELECTED PARAMETERS FOR THE BIOPATH-PROGRAM

U Bergström, A-B Wilkens
Studsvik Energiteknik AB, 1983 06 08

ABSTRACT

A revision of the most necessary input data for the BIOPATH code has been performed. Both nuclide independent and nuclide dependent parameters have been treated.

Especially for the uptake in feed chains statistical properties are given to enable sensitivity analyses of the results.

In appendix the present diet and food consumption habits are reviewed where average intake values are given both for Sweden and for different parts of the world.

ON THE ENVIRONMENTAL IMPACT OF A REPOSITORY FOR SPENT NUCLEAR FUEL

Otto Brotzen, April 1983

SUMMARY

An estimate is presented of the contamination of groundwater discharging at the surface with actinides from spent nuclear fuel emplaced at depth in fractured, water-saturated, crystalline rock. No engineered barriers are considered, and the fuel in this case is free to interact with the groundwater and the host rock. The contaminated groundwater is assumed to reach the surface via a highly water-conducting fracture zone in the rock.

A sequence of processes and factors controlling the final concentration of actinides in the water is considered.

First, oxidative dissolution of spent fuel takes place due to radiolysis of the water. Next, reductive precipitation of uranium, with coprecipitation of the other actinides, results from interaction with reductants in the host rock. The maximum uranium concentration then remaining in solution is about 0.2 microgram per litre. The ratio of other actinides to uranium will reflect the proportions in the fuel.

Only 0.1 per cent or less of the groundwater finally reaching the surface at a local spring or well will come from a properly located repository at depth. The contamination from spent fuel in the groundwater brought to the surface will therefore, after natural mixing en route, amount to 0.0002 microgram of uranium per litre. This uranium is accompanied by a proportional concentration of harmful actinides. Their radiotoxicity is a function of the time between discharge of the fuel from the reactor and their reentry into the surface environment.

Most of this time is spent in transit to the surface from the depth of emplacement. With parallel linear flow in a zone of high conductivity and a retardation of the actinides under reducing conditions, this takes

more than 300 000 years. Radioactive decay during this time reduces the radiotoxic contamination of surfacing groundwater with actinides to a level corresponding to a natural uranium content of 0.05 microgram per litre. This corresponds to less than one per cent of the average concentrations of uranium and radium in water from springs and wells in granitic areas uncontaminated by nuclear installations. Even if instantaneous reentry could take place, the environmental impact of the actinides would only be of the same magnitude as that from natural sources of uranium and radium in the average spring, provided reductive precipitation of uranium has taken place before mixing with shallow groundwater.

ENCAPSULATION OF SPENT NUCLEAR FUEL - SAFETY ANALYSIS

E Söderman
ES-Konsult AB, April 1983

SUMMARY

The radiological safety of the encapsulation procedure planned for the unprocessed nuclear fuel is analysed. Two methods of encapsulation are studied, both including a copper canister. In one process the copper canister with the spent fuel is filled with copper powder and pressed to solid copper metal at high pressure. In the other process lead is cast around the fuel before the canister is sealed by electron beam welding.

The activity decay of the fuel has been going on for 40 years before it arrives to the encapsulation station. This is the basic reason for expecting less activity release and less contamination of the plant than would be the case with fuel recently taken out from the reactors.

In analysing the plant safety, experience from the nuclear power plants, from the planning of the Swedish central storage facility for spent fuel (CLAB) and from La Hague has been used. In those cases where experience is missing, conservative values have been chosen. The analyses is also based on experience of today's technology, although it should be possible to improve the encapsulation process further before time has come to actually build the plant.

The environment activity release will be very low, both at normal operation and following accidents in the plant. Using very conservative release rates also the most severe anticipated accident in the plant will induce a dose to critical group of only 3 Sv.

The staff dose can also be kept low. Due to remote handling, fuel damage will not primarily give staff dose. Of the totally anticipated staff dose of 150 man mSv/year the greatest portion will come from external radiation during repair work in areas where fuel containing canisters by failure cannot be taken away.

The hot isostatic pressed (HIP) canister process contains more operations than does the lead casting and welding procedure. It is therefore expected to give the highest activity release and staff dose unless extra measures are taken to keep them low.

Using remote operation and adequate equipment the encapsulation station with any of the two processes can be built and run with good radiological safety.

FINAL DISPOSAL OF SPENT NUCLEAR - STANDARD PROGRAMME FOR SITE INVESTIGATIONS

Compiled by Ulf Thoregren
Swedish Geological, April 1983

SUMMARY

Like many other countries with similar geological conditions Sweden plans to dispose of its long-lived radioactive nuclear waste by depositing it in final repositories located deep down in the crystalline bedrock.

In order to be able to demonstrate that a given rock formation is suited for waste storage, it is necessary to have knowledge concerning its properties, particularly those that determine groundwater conditions and chemistry within the area. Also of importance are data that shed light on rock mechanics in the area and the occurrence of valuable minerals.

The SKBF/KBS programme includes plans to carry out geological studies of 10-15 areas in different parts of the country during the 1980s. A "standard programme" for these studies is described in the following. The standard programme is intended to serve as a basis for planning of the work and revisions or modifications that may be found to be appropriate in view of local conditions or experience.

FINAL DISPOSAL OF SPENT NUCLEAR - STANDARD PROGRAMME FOR SITE INVESTIGATIONS

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KBS Technical Report No 83-32

FEASIBILITY STUDY OF DETECTION OF DEFECTS IN THICK WELDED COPPER

Tekniska Röntgencentralen AB
April 1983

SUMMARY

The report describes a feasibility study of detecting defects in welded material using focussed ultrasonic beams. The ultrasonic testing has been applied to both electron beam welded thick walled copper containers and copper containers sealed by hot isostatic pressing.

The study shows that focussed ultrasonic technique is capable of detecting small defects in thick welded copper. This is illustrated on test blocks as well as on simulated full scale welds and joints using hot isostatic pressing.

THE INTERACTION OF BENTONITE AND GLASS WITH AQUEOUS MEDIA

M Mosslehi, A Lambrosa, J A Marinsky
State University of New York, Buffalo, NY, USA
April 1983

SUMMARY

It has been demonstrated that Si(OH)_4 is an important constituent of bentonite. In bentonite's natural state the Si(OH)_4 in the bentonite is permeable to the diffusible components of the aqueous medium in contact with it. As a consequence, the pH of the medium is a sensitive function of its ionic strength. Permeability of the Si(OH)_4 to salt is lost, however, by pretreatment of the bentonite with dilute acid to protonate fully the Si(OH)_4 . The pH-producing property of the Si(OH)_4 then is associated with its surface properties.

The glasses (ABS-39 and ABS-Y Marcoule) studied in this research program also contain hydroxylated oxide as an important constituent. These materials, however, are not observed to exhibit gel-like (three-dimensional) properties prior to or after acid pretreatment. Instead they function always as a surface (two-dimensional) and are impermeable to the diffusible components of the aqueous media that come in contact with them. As a result, the pH of the medium is dependent on ionic strength only so far as it affects the counter-ion screening efficiency of the charged surface.

In both systems, bentonite and glass, the pH of the medium is affected by the presence of CO_2 . The lowering of medium pH by CO_2 dissolution results in the neutralization of Si(OH)_4 in the untreated bentonite gel phase or in the surface of the acid pretreated bentonite or glass in contact with the medium. If glass and bentonite are in contact the action of CO_2 at their interface results in both systems striving to reach different equilibria. The result is enhanced chemical reactivity at the interface. To minimize such reactivity CO_2 must be excluded from the atmosphere in contact with the solid-liquid phases.

RADIONUCLIDE DIFFUSION AND MOBILITIES IN COMPACTED BENTONITE

B Torstenfelt, B Allard, K Andersson, H Kipatsi, L Eliasson,
U Olofsson, H Persson
Chalmers University of Technology, 1983 12 15

SUMMARY

Transport rates of the fission products strontium, technetium, iodide and cesium as well as the actinides thorium, protactinium, uranium, neptunium, plutonium and americium in compacted bentonite have been studied.

A sodium bentonite, Wyoming Bentonite MX-80, was used in all experiments, in a few cases mixed with 1% $\text{Fe}_3(\text{PO}_4)_2$, 1% PbO or 0.5% Fe(s), or 1% of one of the minerals apatite, pyrite, chalcopyrite/pyrite, cinnabar or olivine. The aqueous phase was a synthetic groundwater representative of Swedish deep granitic groundwaters, preequilibrated with the clay. Uranium and americium migration was also studied with high carbonate content and 10 mg/l humic acid, respectively, added to the water.

The diffusivity was measured in compacted clay (density $2 \times 10^3 \text{ kg/m}^3$) and calculated from concentration profiles.

The diffusivities for the fission products were of the order of $10^{-12} \text{ m}^2/\text{s}$ under oxidizing conditions and about one order of magnitude lower for technetium under reducing conditions (Fe(s) in the clay).

Penta- and hexavalent actinides had diffusivities around $10^{-13} \text{ m}^2/\text{s}$ and tri- and tetravalent actinides around $10^{-15} \text{ m}^2/\text{s}$. The influence of the additives to the clay or the water was minor in the uranium systems except when Fe(s) was added to the clay, which decreased the transport rate by a factor of 10.

ACTINIDE SOLUTION EQUILIBRIA AND SOLUBILITIES IN GEOLOGIC SYSTEMS

B Allard
Chalmers University of Technology, 1983 04 10

SUMMARY

Stability constants and solubility products for actinide (Th, U, Np, Pu and Am) hydroxide and carbonate complexes have been collected, and total solubilities have been calculated under conditions representative of deep granitic groundwaters (pH 7-9.5; high total carbonate concentration according to $\log(\text{CO}_3) = 0.76\text{pH} - 10.83$; redox potentials corresponding to oxic systems, $E_h \approx 0.8 - 0.06\text{pH}$, and reducing systems, $E_h < 0.3 - 0.06\text{pH}$). The solubility limiting species under reducing systems are $\text{ThO}_2(\text{s})$, $\text{UO}_2(\text{s})$, $\text{NpO}_2(\text{s})$, $\text{PuO}_2(\text{s})$ and $\text{Am}_2(\text{CO}_3)_3(\text{s})$, respectively, and the corresponding calculated maximum solubilities are $3 \times 10^{-10} \text{ M}$ (0.07 $\mu\text{g/l}$) for Th, $0.3 - 250 \times 10^{-10} \text{ M}$ (0.01-6 $\mu\text{g/l}$) for U (depending on the carbonate concentration), $1 \times 10^{-10} \text{ M}$ (0.03 $\mu\text{g/l}$) for Np, $1 - 3 \times 10^{-9} \text{ M}$ (0.2-0.7 $\mu\text{g/l}$) for Pu and $0.6 - 2 \times 10^{-7} \text{ M}$ (15-50 $\mu\text{g/l}$) for Am. The calculated solubilities for U are in fair agreement with observed uranium concentrations in natural deep groundwaters. Hexavalent uranium carbonate species would dominate in solution except under highly reducing conditions ($E_h < 0.2 - 0.06\text{pH}$) and low total carbonate concentrations. Under oxic conditions the concentrations of U and Np would be larger than $10^{-5} - 10^{-6} \text{ M}$ (mg/l-level).

KBS Technical Report No 83-36

IRON CONTENT AND REDUCING CAPACITY OF GRANITES AND BENTONITE

B Torstenfelt, B Allard, W Johansson, T Ittner
Chalmers University of Technology, April 1983

SUMMARY

The iron contents in various granites and in bentonite have been determined. For granites, the content is usually in the range 1-9% (weight) and 2.5-3% for bentonite. Most of the iron is divalent in the granites (70-90%); in bentonite the divalent fraction is 25-50%. A large part of the divalent iron in the granites appears to be accessible for the reduction of dissolved oxygen in an aqueous system.

SURFACE MIGRATION IN SORPTION PROCESSES

A Rasmuson, I Neretnieks
Royal Institute of Technology, March 1983

SUMMARY

Diffusion rates of sorbing chemical species in granites and clays are, in several experiments within the KBS study, higher than can be explained by pore diffusion only. One possible additional transport mechanism is transport of sorbed molecules/ions along the intrapore surfaces.

As a first step a literature investigation on surface migration on solid surfaces has been conducted. A lot of experimental evidence of the mobility of sorbed molecules has been gathered through the years, particularly for metal surfaces and chemical engineering systems. For clays, however, there are only a few articles, and for granites none. Two classes of surface migration models have been proposed in the literature:

- i) Surface flow as a result of a gradient in spreading pressure.
- ii) Surface diffusion as a result of a gradient in concentration.

The surface flow model has only been applied to gaseous systems. However, it should be equally applicable to liquid systems.

The models (i) and (ii) are conceptually very different. However, the resulting expressions for surface flux are complicated and it will not be an easy task to distinguish between them. There seem to be three ways of discriminating between the transport mechanisms:

- a) Temperature dependence
- b) Concentration dependence
- c) Order of magnitude.

EVALUATION OF SOME TRACER TESTS IN THE GRANITIC ROCK AT FINNSJÖN

L Moreno, I Neretnieks, Royal Institute of Technology
C-E Klockars, Swedish Geological
April 1983

SUMMARY

Tracer tests in a granitic rock have been analysed. A nonsorbing tracer - iodide - was used to investigate the hydraulic properties of the pathway. This gives the mean residence time(s) and spreading. The latter may be caused by channeling, hydrodynamic dispersion and interaction with the matrix. Four different models emphasizing different combinations of the various mechanisms were used.

It was found that even the nonsorbing tracer interacts considerably with the matrix. Recent laboratory data which show a high porosity of fissure coating materials indicate that the cause for this interaction may be diffusion of the tracer into the pore water of the matrix.

The residence time(s) and dispersion data obtained from the iodide test were used to predict the behaviour of the sorbing tracer strontium. Independent laboratory data for strontium sorption and diffusion in granite were used in the predictions. The predicted data were in very poor agreement with the field data. The field data were then used to fit the model parameters. The resulting combination of sorption and diffusion data which gives a good fit to the field data was off by a few orders of magnitude.

An examination of some recent sorption and diffusion data show that at the high concentration, which was used in the field test, the sorption isotherm is nonlinear and the sorption capacity considerably smaller than that used in the original predictions. The new data would account for the lower retardation. As, however, the new sorption data have not been performed on the actual fissure coating material in the flowpath and no diffusion measurements at very high concentrations have yet been performed the conclusions are only tentative.

DIFFUSION IN THE MATRIX OF GRANITIC ROCK
FIELD TEST IN THE STRIPA MINE. PART 2

L Birgersson, I Neretnieks
Royal Institute of Technology, March 1983

SUMMARY

A migration experiment in the rock matrix is presented. The experiment has been carried out in "undisturbed" rock, that is in rock under its natural stress environment. Since the experiment was performed at the 360 m-level (in the Stripa mine), the rock was subject to nearly the same conditions as the rock surrounding a nuclear waste repository as proposed in the Swedish concept (KBS).

The results show that all three tracers (Cr-EDTA, Uranine and I^-) have passed the zone disturbed by the presence of the injection hole and migrated into "undisturbed" rock.

These results indicate that it is possible for tracers (and therefore radionuclides) to migrate into the undisturbed rock matrix.

Diffusivities obtained in this experiment are comparable to those obtained in laboratory experiments.

REDOX CONDITIONS IN GROUNDWATERS FROM SVARTBOBERGET, GIDEÅ, FJÄLLVEDEN
AND KAMLUNGE

P Wikberg, I Grenthe, K Axelsen
Royal Institute of Technology, 1983 05 10

SUMMARY

Granitic rock at four different places in Sweden - Svartboberget, Gideå, Fjällveden and Kamlunge - have been investigated in order to find out if they are suitable for the disposal of spent nuclear fuel.

As a part of these investigations groundwaters have been sampled down to depth of about 600 m. Water bearing sections have been isolated with packers, and the water has been pumped up to the surface for the monitoring and sampling. The redox potential E_h was measured with a platinum and a glassy carbon electrode.

The field measurements have been simulated in the laboratory. These experiments have shown that the E_h can be measured under conditions valid for deep ground waters.

The field experiments are complicated by the fact that stable electrode potentials are only slowly obtained. However the measured E_h values show that the ground water at depth in the bedrock is reducing. This picture is confirmed by the low level of uranium present in the waters.

The redox status of the waters has also been calculated using the analytical data obtained from the analysis of sampled water and thermodynamic data for the iron(II)/iron(III) redox couple, and solids of iron(III) oxide hydrate and iron(II) carbonate.

ANALYSIS OF GROUNDWATER FROM DEEP BOREHOLES IN SVARTBOBERGET

Sif Laurent
Swedish Environmental Research Institute, 1983 06 10

SUMMARY

Groundwater from two boreholes in granitic rock at an investigation site in Svartboberget has been sampled and analysed. This is part of a larger program of geological, geophysical and hydrogeological investigations aimed at finding a suitable site for a high level radioactive waste repository.

Four water-bearing levels in each borehole down to the deepest at about 600 m were selected. Prior to sampling, the water-bearing level is isolated between packer sleeves. The water is then pumped to the surface where sensitive parameters such as redox potential, pH, sulphide and oxygen content are measured electrochemically on the flowing water in a system isolated from the air. Water, filter and gas samples are sent to several laboratories for further analysis.

The present report is a presentation of the results of the groundwater analyses. The reliability of the results is discussed but there is no evaluation in relation to geology and hydrogeology. This report presents the basic results from the groundwater analyses to be further evaluated by experts in different fields.

FINAL DISPOSAL OF HIGH-LEVELS WASTE AND SPENT NUCLEAR FUEL -
FOREIGN ACTIVITIES

R Gelin
Studsvik Energiteknik AB, May 1983

SUMMARY

Foreign and international activities on the final disposal of high-level waste and spent nuclear fuel have been reviewed. A considerable research effort is devoted to development of acceptable disposal options. The different technical concepts presently under study are described in the report.

Numerous studies have been made in many countries of the potential risks to future generations from radioactive wastes in underground disposal repositories. In the report the safety assessment studies and existing performance criteria for geological disposal are briefly discussed.

The studies that are being made in Canada, the United States, France and Switzerland are the most interesting for Sweden as these countries also are considering disposal into crystalline rocks.

The overall time-tables in different countries for realisation of the final disposal are rather similar. Normally actual large-scale disposal operations for high-level wastes are not foreseen until after year 2000. In the United States the Congress recently passed the important Nuclear Waste Policy Act. It gives a rather firm timetable for site-selection and construction of nuclear waste disposal facilities. According to this act the first repository for disposal of commercial high-level waste must be in operation not later than January 1998.

FINAL DISPOSAL OF SPENT NUCLEAR FUEL - GEOLOGICAL, HYDROGEOLOGICAL
AND GEOPHYSICAL METHODS FOR SITE CHARACTERIZATION

K Ahlbom, L Carlsson, O Olsson
Swedish Geological, May 1983

SUMMARY

Investigations for the siting of a final repository for high-level radioactive waste are currently being conducted in crystalline rock formations in Sweden. A repository will be located at a depth of about 500 m, and investigations are being carried out in drill holes to below that level.

A standard program has been established for the site investigations, comprising a number of phases:

1. General reconnaissance for selection of study site
2. Detailed investigation on the ground surface
3. Depth investigations in drill holes
4. Evaluation and modelling.

After completion of the reconnaissance studies, which include geological and geophysical reconnaissance measurements and drilling of one deep drill hole, the detailed investigations of the selected site begin. These investigations include surface and depth investigations within an area of approximately 4-8 km².

The surface investigations consist of geophysical measurements including electrical resistivity, magnetization, induced polarization and seismic measurements. Together with geological and tectonic mapping, the surface investigations yield information on the composition and fracturing of the bedrock in the superficial parts of the study sites.

Mapping of the superficial parts of the bedrock are concluded with short percussion and core drillholes down to 150-250 metres in order to determine the dip and character of fracture zones and rock boundaries.

The depth investigations are carried out to characterize the rock at depth from the geological and hydrogeological points of view.

The investigations comprise core drilling to vertical depths of about 600 m, core mapping, geophysical well-logging and different hydraulic downhole measurements.

In core mapping, the emphasis is placed on fracture characterization of the core. The geophysical logging includes three resistivity methods, natural gamma, induced polarization, spontaneous potential and temperature, salinity, pH and Eh of the drill hole fluid. Geophysical measurements are made on the core samples in the laboratory.

The hydraulic measurements include: measurements of hydraulic conductivity by single-hole and cross-hole testing, determination of the hydraulic fracture frequency and determination of groundwater head at different levels in the bedrock.

The single-hole hydraulic tests are performed in 25 m sections as transient tests with injection and fall-off phases. The hydraulic head is calculated from Horner plots and from direct measurement over a long period.

The hydraulic conductivity values obtained are used in descriptive hydraulic models based on geologic-tectonic models of each site under consideration. In the hydraulic models, the bedrock is subdivided into different hydraulic units. The conductivity values obtained in each unit are used to describe the frequency distribution of the conductivity and to calculate an effective hydraulic conductivity versus depth to be used in further numerical groundwater model calculations.

FINAL DISPOSAL OF SPENT NUCLEAR FUEL -
EQUIPMENT FOR SITE CHARACTERIZATION

K Almén, K Hansson, B-E Johansson, G Nilsson, Swedish Geological
O Andersson, IPA-Konsult
P Wikberg, Royal Institute of Technology
H Ahagen, SKBF/KBS
May 1983

ABSTRACT

The suitability of a certain geological formation as a repository for the final disposal of spent nuclear fuel can be determined only after detailed investigation and analysis. The purpose of the investigations is to provide information on the geology and the hydrology and chemistry of the site concerned. The value of these data largely depends on the way in which they have been collected. The report of the findings should enable the investigating party to evaluate the function and the accuracy of the equipment with which field data have been collected for KBS 3.

This report describes the geophysical equipment, the hydraulic testing equipment, the water chemistry sample extracting equipment and the core-logging equipment used. The objectives of the instrument development have been

- to obtain a high data quality
- to collect data automatically in logs and tape recorders for direct transfer to a central processing unit
- to provide back-up in order to counteract loss of data
- to make instrument more efficient.

MODEL CALCULATIONS OF THE GROUNDWATER FLOW AT FINNSJÖN, FJÄLLVEDEN,
GIDEÅ AND KAMLUNGE

L Carlsson, A Winberg, Swedish Geological, Göteborg
B Grundfelt, Kemakta Consultant Company
May 1983

ABSTRACT

Hydraulic properties and conditions were investigated for the study sites of Fjällveden, Gideå, Kamlunge and Svartboberget. Based on geological and tectonical conditions within the sites, the bedrock was divided into different hydraulic units. Within each unit relations between hydraulic conductivity and depth were determined from hydraulic tests performed in 147 to 288 sections in 7 to 16 core-drilled boreholes in each site. The groundwater table was constructed from topographic maps and registration of groundwater head in 23 to 64 boreholes at each site.

Based on the conceptual models of hydraulic units, hydraulic conductivity versus depth and groundwater tables, the groundwater flow conditions were calculated using numerical models. The models used were based on a finite element method in three-dimensions. The three study sites Fjällveden, Gideå and Kamlunge were modelled together with the Finnsjön site. Each site was modelled down to a depth of 1500 m. The results given as groundwater potentials were used to calculate groundwater flow at levels of a potential repository and to estimate trajectories and travel times for groundwater from the repository level.

Calculation of the groundwater recharge and head distribution along profiles coinciding with boreholes made it possible to compare calculated results with measurements on the sites.

USE OF CLAYS AS BUFFERS IN RADIOACTIVE REPOSITORIES

Roland Pusch
University of Luleå, May 25, 1983

SUMMARY

The major physico/mechanical functional criteria which are specified for the three main applications of clay material in repositories, i.e. as canister "overpacks", as "plugs" in boreholes, shafts and tunnels, and as "backfills" in tunnels and shafts, are a low hydraulic conductivity and a swelling ability. The most suitable clay mineral is Na montmorillonite but illite may be accepted under certain circumstances although it does not swell readily. Illite-rich, natural clays are rare but a particular kind of late-glacial sedimentary clay is available in large quantities in Sweden.

Na montmorillonite is the dominant clay mineral in a few bentonites which are exploited commercially, but suitable sources can also be natural Ca bentonites which are converted to the Na state by ion exchange on an industrial scale. Such bentonites are commercially available in bulk or bagged form from several plants in Europe and elsewhere. Refining of illite, on the other hand, requires the establishment of a Swedish plant for the production of sufficient quantities of powdered clay.

For use as canister "overpack", highly compacted bentonite is superior to illite and any reasonably montmorillonite-rich bentonite will do which is not too rich in sulphur. The organic content should be low and heat treatment may be required to bring this content down to an acceptable level. Heating to slightly more than 400°C does not affect the physical properties of neither montmorillonite, nor illite to a significant extent. Bentonite is also very suitable for use as sealing plugs in the form of highly compacted blocks.

For use as backfill in tunnels and shafts, illitic clay is a candidate material which can be compacted on site to the rather high density that is required. Where a swelling capacity is needed, such as in the top part of tunnels, bentonite-based backfills are suitable and if Na saturated clay is used the bentonite fraction can be kept low. Thus, a 10% content of Na bentonite by weight should generally be sufficient for

a well compacted mixture with respect to the required hydraulic conductivity, while a 20-30% content may be needed to arrive at a sufficient swelling power.

The choice of a suitable clay material requires that the substance be properly characterized and tested. It is concluded that rather rigorous analyses are necessary as concerns "over-packs", including mineralogical and granulometrical tests and the determination of the swelling characteristics as well as of certain chemical features. For backfills and for the current checking of all sorts of clay for use as buffer materials, the natural water content, the liquid limit and the swelling ability have to be determined, since they are the fingerprints of this type of soil.

The report ends with a tentative scheme for the possible use of various clay materials in repositories.

STRESS/STRAIN/TIME PROPERTIES OF HIGHLY COMPACTED BENTONITE

Roland Pusch
University of Luleå, May 1983

SUMMARY

The rheological properties of highly compacted bentonite are of practical importance in two respects as concerns the KBS concepts. Thus, they are determinants of the settlement rate of the metal canisters in their deposition holes, and they govern the stress state of the canisters in the event of rock displacements.

The rather comprehensive research work in recent years on the physico-chemical nature of smectite-rich clays, points to the importance of the strongly mineral-adsorbed water for their bulk behavior. The impact of shear stresses on the microstructure is the key problem, which requires the formulation of a reasonable physical model as a basis of the derivation of mathematical analogies for time-dependent stress/strain relationships. In this paper, a recently developed creep theory based on statistical mechanics has been used to analyze a number of experimental creep curves, the conclusion being that the creep behavior of dense MX-80 bentonite is in agreement with the physical model, and that the average bond strength is within the hydrogen bond region. The latter conclusion thus indicates that interparticle displacements leading to macroscopic creep takes place in interparticle and intraparticle water lattices. These findings were taken as a justification to apply the creep theory to a prediction of the settlement over a one million year period. It gave an estimated settlement of 1 cm at maximum, which is of no practical significance.

The thixotropic and viscous properties of highly compacted bentonite present certain difficulties in the determination and evaluation of the stress/strain/time parameters that are required for ordinary elastic and elasto-plastic analyses. Still, these parameters could be sufficiently well identified to allow for a preliminary estimation of the stresses induced in the metal canisters by slight rock displacements. The analysis, which needs to be deepened in order to get a more realistic picture of the impact of such displacements on the physical state of the canisters, suggests that a 1 cm rapid shear perpendicular to the axes of the canisters can take place without harming them.

MODEL CALCULATIONS OF THE MIGRATION OF RADIO-NUCLIDES FROM A
REPOSITORY FOR SPENT NUCLEAR FUEL

A Bengtsson, Kemakta Consultant Company
M Magnusson, I Neretnieks, A Rasmuson, Royal Institute of Technology
May 1983

SUMMARY

Release and migration calculations for all important radionuclides which potentially could escape from a repository for spent nuclear fuel have been performed. The calculations have been done for a repository in fissured crystalline rock with models which account for the leaching of the nuclides from the fuel due to dissolution in the moving water and the further transport along the fissures of the rock. Radionuclide sorption retarding the nuclides is accounted for by the concept of matrix diffusion and sorption on the microfissure surfaces. Dispersion and channeling are modeled as Fickian dispersion. Chain decay is accounted for. Calculations have been performed for migration distances of 5, 50 and 100 m. Sorption data and solubility data for oxidizing as well as reducing conditions have been used.

Transport by sorption on colloidal matter in the water is also treated.

In the "central" case the leaching starts after 100,000 years due to the long life time of the canister, but calculations have also been made for a case of some defective canisters where the leaching starts after 100 years. The results are presented as release rates to the biosphere at the various times.

KBS Technical Report No 83-49

DOSE AND DOSE COMMITMENT CALCULATIONS FROM GROUNDWATERBORNE RADIOACTIVE ELEMENTS RELEASED FROM A REPOSITORY FOR SPENT NUCLEAR FUEL

U Bergström
Studsvik Energiteknik AB, May 1983

ABSTRACT

The turnover of radioactive matter entering the biosphere with groundwater has been studied with regard to exposure and doses to critical groups and populations. Two main recipients, a well and a lake, have been considered for the inflow of groundwaterborne nuclides.

Mathematical models of a set of coupled ecosystems on regional, intermediate and global levels have been used for calculations of doses. The intermediate system refers to the Baltic Sea.

The mathematical treatment of the model is based upon compartment theory with first order kinetics and also includes products in decay chains.

The time-dependent exposures have been studied for certain long-lived nuclides of radiological interest in waste from disposed fuel. Dose and dose commitment have been calculated for different episodes for inflow to the biosphere.

CALCULATION OF FLUXES THROUGH A REPOSITORY CAUSED BY A LOCAL WELL

R Thunvik
Royal Institute of Technology, May 1983

ABSTRACT

The purpose of the present study is to roughly estimate the flux through a radioactive repository in relation to the flux into a local well under various conditions. The well is assumed to be located at a depth of either 60 or 200 metres below the ground surface. Two main settings are considered, in one the well is located above the centre of the repository, and in the other it is located in a vertical fracture zone at a distance of 100 metres from one of the outer edges of the repository. It is assumed in the calculations that the well is the only cause of the groundwater flow. The withdrawal from the well is assumed to be $6 \text{ m}^3/\text{day}$.

The flow domain is characterized by a rather low permeability, which is assumed to decrease with depth over the flow domain. The boundary conditions considered are either a continuously saturated upper boundary and impervious lateral boundaries, or a phreatic upper boundary and hydrostatic lateral boundaries. In all examples an impervious bottom is assumed to be located at a depth of 1500 metres below the ground surface, and the repository is assumed to be located at a depth of 500 metres. The areal extent of the repository is 0.8 km^2 in the first setting and 1 km^2 in the second one. The areal extent of the flow domain is 1.1 km^2 in the first setting and 12 or 24 km^2 in the second one.

The ratio of the flux through the repository to the flux into the well was obtained to be in the range from about 10^{-5} to 10^{-3} , depending on the boundary conditions and the depth of the well. The lowest figures were obtained in the examples, in which the upper boundary was assumed to be continually saturated. It is concluded that these examples may be considered representative of the actual flow problem. This conclusion is based upon the fact that in the case of a phreatic boundary the drawdown caused by the well was very small and the flow responses were very slow, implying that rather a small infiltration rate is required to maintain saturated conditions at the upper boundary. The regional gradients caused by the well were rather small in comparison with the typically naturally occurring gradients. The flow to the well will therefore have little influence on the regional flow pattern in most practical cases.

GWHRT - A FINITE ELEMENT SOLUTION TO THE COUPLED GROUND WATER FLOW AND HEAT TRANSPORT PROBLEM IN THREE DIMENSIONS

B Grundfelt
Kemakta Consultant Company, May 1983

SUMMARY

A program package for modelling group water flow with the Finite Element Method (FEM) is presented. The package, that comprises programs for pre- and postprocessing data for the FEM-model, has been extensively used together with the GWHRT-model developed by Roger Thunvik.

Thunvik's hydraulic model is one-, two- or threedimensional. It solves the ground water pressure distribution and the temperature distribution in the modelled domain using the Finite Element Method. Also saturated/unsaturated flow can be modelled.

The calculations performed in the KBS-3 study are three-dimensional steady state calculations for isothermal conditions. Such a calculation typically involves the following steps:

- Generation of the finite element mesh.
- Assigning boundary conditions and material properties to the domain.
- Solving for the hydraulic pressure distribution.
- Checking the results.
- Presenting the results.

In the program package HYPAC these steps are implemented. This report deals mainly with the type of calculations made in the KBS-3 study although the program package in principal can handle also other types of calculations like for instance transient saturated-unsaturated flow.

EVALUATION OF THE GEOLOGICAL, GEOPHYSICAL AND HYDROGEOLOGICAL CONDITIONS
AT FJÄLLVEDEN

K Ahlbom, L Carlsson, L-E Carlsten, O Duran, N-Å Larsson, O Olsson
Swedish Geological, May 1983

SUMMARY

The Fjällveden study site has a flat topography and a high percentage of outcrops. The main type of rock in the area is veined gneiss with a north-east structural strike and vertical dip. The veined gneiss contains sulphide minerals, primarily pyrite and pyrrhotite, in the form of fracture minerals and as impregnations. The concentrations of economically valuable minerals are so small that mining in the area is not realistic. In conformity with the gneiss structure there are strata of granite gneiss. They have a mean width of 3 m and occur at a mean distance of 100 m. These strata are probably of greater continuity in the horizontal direction than in the vertical. The proportion of granite gneiss in the drill holes is 3 %.

The rock mass has a fracture frequency of 4.0 fractures per metre within the upper 100 metres. The frequency decreases with increasing depth and below the 300 m level it is 1.8 fractures per metre. The proportion of hydraulically conductive fractures is lower than the total fracture frequency.

The Fjällveden study site is delimited to the north-east and south-west by regional fracture zones, 80-90 m wide and dipping approx 75° towards the south-west. The fracture zones contain wide sections of crushed and clay-altered rock, mylonites and breccias. These zones are 1250 million years old or older.

The regional zones delimit a block at least 3 x 3 km large. This block contains only local fracture zones. Drill hole examinations indicate that the fracture zones are small and of a mean width of 5 m. They are steep and contain minor sections of crushed and clay-altered rock. Within the upper 100-200 metres horizontal fractures can be found. Below this level no horizontal fracture zones have been observed. Common fracture minerals in fracture zones are calcite, kaolinite, chlorite and illite. The extensive presence of kaolinite in the fracture zones and the bedrock as a whole indicates that Fjällveden is or has been subject to deeply extending weathering.

Existing granite gneiss strata possess higher hydraulic conductivity than the surrounding veined gneiss. At a depth of 500 m the granite gneiss has a hydraulic conductivity of 3×10^{-9} m/s, the corresponding value in the surrounding bedrock being 2×10^{-11} m/s. This gives the rock mass anisotropic hydraulic properties with a higher hydraulic conductivity in the north-easterly and vertical directions and a lower conductivity perpendicular to this.

The hydraulic conductivity in the veined gneiss and the granite gneiss decreases substantially with increasing depth. For the rock mass as a whole, i.e. by forming the average of all data from both rock types, the hydraulic conductivity decreases from approx 10^{-8} m/s at the surface rock to approx 10^{-11} m/s at a depth of 600 m.

The hydraulic conductivity in the local fracture zones in the Fjällveden area is 5×10^{-9} m/s at the 500 m level. In the case of fracture zones the hydraulic conductivity decreases with increasing depth in the same way as that of the rock mass. An interesting fact is that the hydraulic conductivity of the fracture zones is lower than in the granite gneiss.

The flat topography of the area implies that the hydraulic gradients in the bedrock are small. This has also been recorded by means of piezometric measurements. Locally, there are low water pressures within granite gneiss strata at the 600 m depth level in the south-western part of the area. These strata are probably in hydraulic contact with the regionally delimiting fracture zone to the south-west.

EVALUATION OF THE GEOLOGICAL, GEOPHYSICAL AND HYDROGEOLOGICAL CONDITIONS AT GIDEÅ

K Ahlbom, B Albino, L Carlsson, G Nilsson, O Olsson, L Stenberg, H Timje
Swedish Geological, May 1983

SUMMARY

The Gideå study site has a flat topography, insignificant soil depth and a high percentage of outcrops. The dominating rock type in the area is veined gneiss of north-east structural strike and small dip. The veined gneiss contains sulphide minerals, primarily pyrite and pyrrhotite in the form of impregnations or as minor enrichments parallel with the gneiss structure. Economically valuable minerals are present in so small quantities that mining in the area is not realistic.

In conformity with the structure of the gneiss there are strata or irregular bodies of granite gneiss. The proportion of granite gneiss in the drill holes is 6%.

The fracture frequency in the rock mass is more than 4.0 fractures per metre down to a depth of 400 m. Below the 500 m level, the fracture frequency is 2.0 fractures per metre. Outside the Gideå study site there are regional fracture zones towards the west-north-west and the north-west. These zones are 50-100 m wide, or more, and dip vertically. The regional zones delimit a plateau with an area of c. 100 km².

Within the study site there are defined local fracture zones at a mutual distance of 400-800 m. These local fracture zones delimit a triangular block of rock with a top surface of 1.8 km². This block is transversed by two local fracture zones, approx 4 m wide. Drill hole investigations indicate that the fracture zones in the study site have a mean width of c. 11 m and contain small portions of crushed and clay-altered rock. The fracture zones are steeply dipping with the exception of two sub-horizontal fracture zones in the northern and eastern parts of the area which are dipping outwards from the study site. Horizontal fractures of vast lateral extent can be found in the upper 100-200 metres. Under this level no horizontal fracture zones have been observed. Frequently occurring fracture minerals in the fracture zones are calcite, kaolinite, chlorite, laumontite, pyrite and the clay minerals illite and smectite.

Existing strata or bodies of granite gneiss have a higher hydraulic conductivity than the surrounding veined gneiss. At a depth of 500 m the

granite gneiss has a hydraulic conductivity of 1.5×10^{-10} m/s, that of the surrounding bedrock being 2×10^{-11} m/s. This implies anisotropic hydraulic properties in the rock mass with a higher hydraulic conductivity horizontally.

The hydraulic conductivity in the veined gneiss and granite gneiss decreases substantially with increasing depth. In the rock mass as a whole, i.e. by forming the average of all data from both rock types, the hydraulic conductivity decreases from c. 4×10^{-9} m/s at 100 m depth to c. 2×10^{-11} m/s at a depth of 500 m.

The hydraulic conductivity in the local fracture zones at Gideå is 9×10^{-11} m/s at a depth of 500 m. The hydraulic conductivity in the fracture zones decreases with increasing depth in the same way as that of the rock mass. An interesting fact is that the hydraulic conductivity of the fracture zones is lower than in the granite gneiss.

The flat topography of the area implies that the hydraulic gradients in the bedrock are small. This has also been recorded by means of piezometric measurements.

EVALUATION OF THE GEOLOGICAL, GEOPHYSICAL AND HYDROGEOLOGICAL CONDITIONS AT KAMLUNGE

K Ahlbom, B Albino, L Carlsson, J Danielsson, G Nilsson, O Olsson,
S Sehlstedt, V Stejskal, L Stenberg
Swedish Geological, May 1983

SUMMARY

The Kamlunge study site constitutes a 16 km² mountain plateau. The topography of the plateau is flat, the soil cover is thin, and in the western part, there is a high percentage of outcrops. The most commonly occurring rock types are gneisses and red granite (Lina granite). A rock type with granodioritic to dioritic composition also occurs. Concentrations of economically valuable minerals are so small that mining operations are not feasible in the area.

The rock mass exhibits a fracture frequency of more than 4.0 fractures per metre down to a depth of 200 metres. Below 300 metres, the fracture frequency is approximately 2.0 fractures per metre.

The Kamlunge study site is surrounded by regional fracture zones to the north, east and west delimiting a 16 km² triangular block. The regional zone to the west of the study site has a width of about 550 m.

Only local fracture zones spaced 500-1500 m apart occur within the study site. The local fracture zones are generally steeply inclined and strike to the north-west and the north-east. At a depth of 555 m below Kamlungekölen, a horizontal fracture zone has been encountered in 4 of the deep drill holes. This fracture zone is permeable to water but less crushed and weathered than the steeply inclined fracture zones. Moreover, horizontal fractures of large lateral extent can occur in the upper 100-200 metres. Common fracture minerals in the fracture zones are calcite, chlorite, laumontite, smectite and various types of iron oxides.

The hydraulic conductivity of the rock mass decreases markedly with depth. It decreases from about 2×10^{-9} m/s at a depth of 100 metres to about 10^{-11} m/s at a depth of 500 metres.

The hydraulic conductivity of the local fracture zones at Kamlunge is 7×10^{-10} m/s at a depth of 500 m. The hydraulic conductivity decreases with depth more slowly in the fracture zones than in the rock mass. The large hydraulic gradients found on the margins of the Kamlungekölen do not affect the groundwater flows at a depth of 500 m within the study site.

EVALUATION OF THE GEOLOGICAL, GEOPHYSICAL AND HYDROGEOLOGICAL CONDITIONS
AT SVARTBOBERGET

K Ahlbom, L Carlsson, B Gentschein, A Jämtlid, O Olsson, S Tirén
Swedish Geological, May 1983

SUMMARY

The Svartboberget study site is an elongate hill, approx 2.5 km wide and 5 km long. The difference in altitude between the upper part of Svartboberget and the adjacent valleys to the northeast and southwest is 85 and 75 m, respectively. The depth of the Quaternary sedimentary cover is small and the percentage of exposed rock is small. The main rock type is a migmatitic paragneiss with a great amount of neosome, granite, and subordinate veins. In the northern part of the area there is a 150 m thick layer of graphitic gneiss. The content of economic minerals within this gneiss is small but future mining could not be excluded.

The fracture frequency of the rock mass is 2.6 fractures per metre and the variation with depth is insignificant.

The Svartboberget study site is situated between two north-northwest orientated valleys which are defined by regional fracture zones. The thickness of these zones are from 10 to more than 250 m. They dip 30 to 40 degrees towards the southwest and can be traced over long distances. The fracture zones consist of discrete sections, locally more than 30 m thick, of highly fractured and subordinate crushed rock. These sections are separate from each other by c. 10-125 m wide sections of normally fractured rock.

The regional fracture zones delimit a rock plinth, approx 5 km² large, consisting of the actual hill Svartboberget. Within this plinth there are local fracture zones with a mutual distance of 100-500 m. In all, 17 local fracture zones with varying dip have been located. These fracture zones intersect each other at different depths. Drill hole investigations indicate that the fracture zones have a mean width of 16 m. They contain minor parts of crushed rock. Common infilling in the fracture zones are calcite, chlorite, illite and zeolite.

The hydraulic conductivity in the rock mass of Svartboberget decreases with depth. At 100 m depth, the conductivity is c. 4×10^{-9} m/s and at 500 m c. 5×10^{-11} m/s. The hydraulic conductivity of the local fracture zones in Svartboberget is at 500 m depth 8×10^{-10} m/s. In the fracture zones the hydraulic conductivity decreases with depth in a corresponding way.

The topographic relief of the area implies that there is a high hydraulic gradient in the bedrock. This has also been confirmed by piezometric measurements where i.a. pressure differences of a 30 m water column have been registered in a drill hole.

I: EVALUATION OF THE HYDROGEOLOGICAL CONDITIONS AT FINNSJÖN
L Carlsson, G Gidlund

II: SUPPLEMENTARY GEOPHYSICAL INVESTIGATIONS OF THE STÄRNÖ PENINSULA
B Hesselström

Swedish Geological, May 1983

SUMMARY

Within the study area Finnsjön in northern Uppland, hydrogeological and geological investigations were carried out during 1977-1982. The object of the investigations was to obtain a better knowledge of the groundwater occurrences and movement in crystalline bedrock and to test different investigation methods.

The study area is about 25 km² and has a flat topography due to the Sub-Cambrian peneplain. The differences in altitude are in general less than 15 m. The bedrock is composed of metamorphic sediments and vulcanites of Sweco-Karelian age. The dominating rock type is granite gneiss. The area is covered to 85% by Quaternary deposits, mainly till. Eight core drill holes and seventeen percussion drill holes were made in the area. The cores were mapped regarding rock type, fracture fillings and fracture frequency. In the core drill holes water injection tests were performed in sealed-off two- or three-meter sections. The groundwater head was measured in open boreholes and the sealed-off sections in the drill holes.

The tectonics in the area were studied from satellite and aerial photographs. The fracture frequency measured on drill cores is an average 2.5 fractures per meter. No tendency of decreasing frequency with depth was obtained.

The hydrometeorological conditions of the area were obtained by measurements of i.a. precipitation and run-off. Calculations and field-mapping showed that about 30% of the study area consists of areas for groundwater discharge. The precipitation is on the average 670 mm per year and the run-off about 240 mm per year.

The bedrock was divided into different hydraulic units with different hydraulic properties. The division was based on tectonic considerations and results from drillings and hydraulic tests. Depending on the scale used, a different degree of accuracy was applied in the division of the units. In principle the bedrock was subdivided in fracture zones and rock mass.

The hydraulic conductivity within each unit was calculated from the statistical frequency distribution of the values obtained from the field tests. A decrease of conductivity versus depth was observed. However, this decrease was strongly influenced by the measurement limit.

NEOTECTONICS IN NORTHERN SWEDEN - GEOPHYSICAL INVESTIGATIONS

L Henkel, K Hult, L Eriksson, Geological Survey of Sweden
L Johansson, Swedish Geological, May 1983

ABSTRACT

The report treats the geophysical aspects of the late quaternary faults in the Norrbotten district, and the connection between these faults and older fracture zones. The older fracture zones are mapped out mainly by aeromagnetic interpretation.

Three late quaternary fault complexes are included, namely the Lansjärv fault, the Kärkejaure fault and the Pärvie fault.

Around the Lansjärv fault, older regional and local fault systems have been mapped out by aeromagnetic interpretation. The interpretation in the vicinity of the fault was made from high resolution aeromagnetic data, augmented with ground geophysical surveys. The technique and the limitations of the aeromagnetic interpretation are discussed.

Maps and trend distribution diagrams of the older fault zones are presented, together with interpretation of the displacements at some of these zones.

It has been found that the late quaternary faults at Lansjärv are largely influenced by the older fault zones, but that fracturing outside of these zones is quite common. It is also suspected that recent movements may have occurred within major older fault zones, but escaped detection in these places.

Possible mechanisms, i.e. post-glacial uplift and plate tectonic motion, of the late quaternary fault movements are preliminarily discussed.

Also around the Kärkejaure fault, aeromagnetic interpretation from high-resolution data has been made. This has revealed that the fault entirely follows an older fault zone, that previously has escaped detection. It has also been found that the late quaternary fault has been influenced by the local fracture system belonging to the Kärkejaure gabbro.

At the Lansjärv fault, also seismic refraction investigation has been made. The results indicate that late quaternary block motion has occurred at several events. The throw of the fault is estimated from the thickness of weathered rock, preserved on the downthrown side.

Airborne electromagnetic (RAMA) surveys over late quaternary faults are discussed and compared with ground electromagnetic methods. It is shown that the late quaternary faults generally are represented by conductive zones in the bedrock, i.e. water-bearing fracture zones. A case of Slingram survey over a late quaternary fault, where it is shown that the recent fault apparently has engaged the central part of an older fracture zone, is presented.

In the Tjärrojåkka and Aitejåkka areas, where ground geophysical data from earlier ore prospecting are available, these have been used, together with supplementary surveys, for the investigation of the faults of the Pärvie complex. It has been found that at Tjärrojåkka, the Pärvie fault coincides with an older tectonic zone. The seismic survey undertaken indicates that substantial preglacial displacement has occurred at the zone.

Also the fault at Aitejåkka has been found to follow an older tectonic zone.

NEOTECTONICS IN NORTHERN SWEDEN - GEOLOGICAL INVESTIGATIONS

R Lagerbäck, F Witschard
Geological Survey of Sweden, May 1983

ABSTRACT

Fairly large areas around the formerly known quaternary faults have been air photo interpreted. The former picture of the faults has thus been complemented in a few significant respects. The fault known as the Pärvie fault has been found to extend somewhat further towards the south, thereby crossing the valley of the Stora Lule river. Furthermore, another fault has been discovered in the Lansjärv region, and thus the faults in this area form a better fit to the regional pattern, with a SSW - NNE trend and a relative uplift of the eastern part.

The fault scarps have been leveled photogrammetrically, and reproduced on maps on the scales of 1:50 000 and 1:100 000, and on overview maps on the scale of 1:250 000. The highest leveled scarps somewhat exceed 30 m. The total length of the faults is roughly 300 km.

During the air photo interpretation, several landslides have been detected, and it seems evident from their location that there is a causal connection between faults and landslides.

As far as possible, the age of the faults relative to the deglaciation has been estimated. The former opinion, that the faults have formed in connection with the deglaciation, has been supported. However, it seems evident that the different faults are not simultaneously formed, but created at separate events. This may also be the case for the different parts of the faults.

Most of the outcrops along the fault zones have been visited in the field, and investigated regarding rock types and tectonic influence. Representative samples have been collected, and thin sections of these investigated under the microscope. Often, the bedrock shows signs of older tectonic influence, and it seems that the faults largely have been released along existing zones of weakness in the bedrock. However, striking exceptions, with fracturing through unaltered rock, have been found in several places.

The faults illustrated in the maps below undoubtedly represent the most important signs of late quaternary fault activity in Norrbotten east of

the Caledonian mountains. Further and more exhaustive investigation will hardly change this perception in any essential way. Certainly, minor faults of small scarp height may have escaped detection, but it is unlikely that any major faults have been overlooked.

Neither will the picture of the distribution of the landslides change significantly. The geographic, and very probably also causal connection between faults and landslides seems obvious in both Finland and Sweden.

The matter of the geographic distribution, appearance, and age of the faults may for the time being be regarded as sufficiently illustrated, and it is about time that other aspects of the phenomenon are dealt with. Among these are the connection between the shape of the faults and the qualities of the bedrock, petrographic as well as physical. We need a satisfactory tectonic explanation for the faults. Undoubtedly, the glacial - isostatic forces have a central role, but in what way, and why just in this region? What role do plate-tectonic processes play? The strike of the faults is approximately perpendicular to the direction of plate motion, and compressive forces have acted at the formation of the faults.

To settle these matters, further work is required, including documentation and compilation of the pre-Quaternary geology, extended and detailed aeromagnetic interpretation, reconstruction of the process of deglaciation, tectonic analysis, and model computation. The essential work encompasses a wide variety of sciences and, to be successful, the further investigation must be conducted in a truly interdisciplinary spirit.

CHEMISTRY OF DEEP GROUNDWATERS FROM GRANITIC BEDROCK

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SÅ Larson, E-L Tullborg, Swedish Geological
P Wikberg, Royal Institute of Technology
May 1983

SUMMARY

Water analysis data from Fjällveden, Gideå, Svartboberget and Kamlunge (7 different drilling holes, 26 sampling levels at vertical depths between 100 and 600 m) are discussed. Most of the waters are Na^+ - Ca^{2+} - HCO_3^- -dominated with a total salt content of 200-300 mg/l and pH of 8-9. Intrusions of Na^+ - Cl^- -dominated saline waters (up to 650 mg/l) are observed at great depth. The conditions are generally strongly reducing. The presence of clayish material (kaolinite, smectites) and zeolites in the fractures appears to have a large influence on the cation concentration ratios. The contents of organics, largely fulvic acids of intermediate molecular weight (<1000) is considerable (1-5 mg/l). Measurements on stable isotopes (^{18}O and deuterium) indicate a non-marine origin of the water and only minor exchanges with the surroundings. The presence of tritium is evidence of intrusions of young waters in some of the samples, probably due to the disturbances during drilling and sampling.

ON THE SOLUBILITY OF TECHNETIUM IN GEOCHEMICAL SYSTEMS

B Allard, B Torstenfelt,
Chalmers University of Technology, 1983 05 05

SUMMARY

The solubility of technetium under groundwater conditions has been calculated from hydrolysis and redox data.

Under reducing conditions ($E_h < 0.27-0.06\text{pH}$, V) a formation of elementary Tc(s) is feasible. Total concentrations in solution may go down to the 10^{-9} M-level or below.

Under mildly reducing conditions (E_h between $0.27-0.06\text{pH}$ and $0.45-0.06\text{pH}$) a constant solubility of the order 10^{-5} M would be achieved, limited by $\text{TcO}_2(\text{s})$ and with $\text{TcO}(\text{OH})_2$ as dominating species in solution.

At higher redox potentials ($E_h > 0.5-0.06\text{pH}$) the heptavalent TcO_4^- dominates entirely, and no solubility limiting phase would be expected in most natural waters.

SORPTION BEHAVIOUR OF WELL-DEFINED OXIDATION STATES

B Allard, U Olofsson, B Torstenfelt, H Kipatsi
Chalmers University of Technology, 1983 05 15

SUMMARY

The sorption of the actinides Am(III), Th(IV), Np(V), Pa(V), U(VI) and Pu has been studied as a function of pH (2-12) for two nuclide concentrations (10^{-7} - 10^{-9} M) (only one for Pa and U) in the systems Al_2O_3 - 0.01 M NaClO_4 and SiO_2 - 0.01 M NaClO_4 . Distribution coefficients have been determined by a batch technique after various contact times (6h - 6w) at constant temperature (25°C) in systems equilibrated with air. The observed sorption behaviour indicates a predominantly physical adsorption mechanism, where pH of the aqueous phase is the principal chemical parameter of influence. The sorption is highly related to the degree of hydrolysis, with a maximum in the pH-region where neutral species dominate and with a reduction of the sorption under conditions when anionic species (hydroxides or carbonates) would exist in solution. This is particularly the case for U(VI) at pH above 7-8 when anionic carbonate complexes would be formed. Plutonium is predominantly tetravalent under the present conditions, as indicated by the sorption behaviour.

KBS Technical Report No 83-62

THE DISTRIBUTION COEFFICIENT CONCEPT AND ASPECTS ON EXPERIMENTAL
DISTRIBUTION

B Allard, K Andersson, B Torstenfelt
Chalmers University of Technology, May 1983

SUMMARY

Aspects on the distribution coefficient concept, sorption mechanisms and experimental studies of sorption phenomena are given.

(The report is only an introductory summary, where only a few aspects of the topic are given. A full revised Technical Report with a detailed discussion of sorption phenomena etc is in progress. This report will be issued in the KBS TR-series during 1984.)

SORPTION OF RADIONUCLIDES IN GEOLOGIC SYSTEMS

K Andersson, B Torstenfelt, B Allard
Chalmers University of Technology, 1983 06 15

SUMMARY

The sorption of radionuclides (cesium, strontium, iodide, technetium and carbon-14) has been studied as a function of pH, nuclide concentration, ionic strength and contact time on geologic material in distribution experiments (batch-wise equilibrium studies and transport studies in columns).

Important parameters governing the interaction of radionuclides with geologic materials are pH, the ionic strength and the amount of complex-forming agents in the aqueous phase, and the mineralogy of the geologic media, the cation exchange capacity of the corresponding minerals, the surface to mass ratio and the amount of complex-forming agents in the solid phase.

Three main sorption mechanisms have been exemplified: Electrostatic interactions (ion exchange processes), which depend on the exchange capacity of the solid, the pH and ionic strength of the water and concentration of the sorbing species; chemisorption processes, where complex-forming agents, either in solution or on the solid phase interact with the radionuclide; physical adsorption phenomena, where cationic and neutral complexes are sorbed on solid surfaces by non-specific forces.

The sorption mechanism for cesium and strontium is mainly ion exchange and for iodide, technetium and carbon (as carbonate) chemisorption under oxic conditions. Technetium in the reduced form (as technetium(IV)hydroxide/oxide) sorbs like the actinides mainly by physical adsorption.

Radionuclides sorbing by ion exchange mechanisms are found to migrate in rock through microfissures and high-capacity minerals. A similar migration was not found for radionuclides sorbing mainly by chemisorption or physical adsorption processes.

ION EXCHANGE CAPACITIES AND SURFACE AREAS OF SOME MAJOR COMPONENTS AND
COMMON FRACTURE FILLING MATERIALS OF IGNEOUS ROCKS

B Allard, M Karlsson, Chalmers University of Technology
E-L Tullborg, S A Larson, Swedish Geological
May 1983

SUMMARY

Surface/mass ratios have been determined by the ethylene glycol method and the BET-method, and an- and cation exchange capacities vs pH have been measured by an isotopic dilution technique for some 30-35 pure minerals. These minerals, which are representative of major components and common accessory minerals, including fracture minerals, of crystalline rocks, have also been used in radionuclide sorption studies within the Swedish waste program.

Generally, the anion exchange capacities are low (<1 meq/kg) for all the minerals. The cation exchange capacities, which increase with pH, are ≤ 1 meq/kg for the non-silicates with few exceptions, 1-10 meq/kg for most of the silicates except the phyllosilicates and ranging from 10 up to 800 meq/kg for clay minerals (e.g. montmorillonite) and some of the zeolites (e.g. stilbite).

SORPTION OF ACTINIDES ON URANIUM DIOXIDE AND ZIRCONIUM DIOXIDE IN
CONNECTION WITH LEACHING OF URANIUM DIOXIDE FUEL

B Allard, N Berner, K Andersson, U Olofsson, B Torstenfelt
Chalmers University of Technology
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May 1983

SUMMARY

The sorption of Cs, Sr, U(VI), Np(V), Pu and Am(III) on UO_2 (fuel pellets) and ZrO_2 has been studied by a batch technique. The aqueous phase has been an artificial groundwater (pH 8, total carbonate ca 2 mM) or a salt solution (0.01 M $NaClO_4$, variation of pH between 2 and 10). Comparison is made with the sorption behaviour on other oxides such as magnetite, alumina and quartz.

The most important chemical parameters affecting the sorption behaviour of the actinides on the studied oxides are pH and the presence of carbonate (for U(VI) in particular). The uptake of Cs and Sr is poor on all of the oxides under the present conditions.

THE MOVEMENT OF RADIONUCLIDES PAST A REDOX FRONT

I Neretnieks, B Aslund
Royal Institute of Technology, 1983 04 22

SUMMARY

It is assumed that radiolysis of water in a penetrated canister containing spent fuel has occurred. Radionuclides and oxidizing agents are diffusing from the corroded canister and out through the clay barrier. A concentration front of radionuclides as well as of oxidizing agents is developed in the water that is flowing past the repository. The fronts propagate in a plane fissure with infinite extension. In the undisturbed bedrock reducing conditions normally prevail. The oxidizing agents are consumed by oxidation of the reducing components of the bedrock (primarily Fe(II)), and an oxidized region is developed. A redox front develops between the oxidizing and reducing regions. Some of the nuclides are much more soluble in an oxidizing environment than in a reducing. These nuclides will precipitate when they reach the redox front. The redox front moves much slower than the nuclides and can be assumed to be stationary at every moment.

The movement of the radionuclides in the flowing water up to and past the redox front have been computed. In the computations the transverse diffusion and precipitation at the redox front are accounted for. That part of the nuclides which is not precipitated at the passage of the redox front has been computed for different solubility ratios and at various distances downstream from the canister. The solubility ratio indicates the solubility of a nuclide in a reducing environment divided by the solubility in an oxidizing environment.

SOME NOTES IN CONNECTION WITH THE STUDIES OF FINAL DISPOSAL OF SPENT FUEL. PART 2.

I Neretnieks
Royal Institute of Technology, May 1983

SUMMARY

This report contains the following "notes" on various subjects related to the Swedish KBS III study.

Neretnieks I.

A note on the transport of hydrogen from a waste canister.

Andersson L., Neretnieks I., Rasmuson A.

Water flow in bedrock. Estimation of influence of transmissive shaft and borehole.

Rasmuson A., Neretnieks I.

A note on the impact of transverse dispersion and "immediately" dissolved radionuclides.

Andersson L., Neretnieks I., Rasmuson A.

Some simple calculations of potential fields in rock mass underneath two typified topographies.

TWO DIMENSIONAL MOVEMENTS OF A REDOX FRONT DOWNSTREAM FROM A REPOSITORY FOR NUCLEAR WASTE

I Neretnieks, B Aslund
Royal Institute of Technology, 1983 06 03

SUMMARY

A broken canister with radioactive waste from nuclear power plants is assumed to have come in contact with water. Radiation (mainly α) will radiolyze the water to form the main products: hydrogen and hydrogen peroxide. The rate of the radiolysis decreases with time. The hydrogen is not very reactive and will diffuse away. The hydrogen peroxide will oxidize ferrous iron in the backfill and rock matrix to ferric iron. Normally there is a reducing environment in the bedrock, but with the migration of the hydrogen peroxide into the bedrock it will change to an oxidizing environment. The hydrogen peroxide is very reactive and cannot coexist with ferrous iron. Thus a sharp front between oxidizing and reducing conditions is developed. This redox front moves very slowly in comparison with the movement of the radionuclides in the fissure. Some of the nuclides (Np, Tc and U) have a much lower solubility under reducing conditions than under oxidizing conditions. These will precipitate at the redox front to the solubility concentration.

A model with diffusion in the axial and transverse directions, advection in the axial direction and diffusion into the rock matrix is used in a numerical computation to describe the movement of the redox front. An analytical solution with advection only in the fissure has also been used.

Christensen and Bjergbakke (1982b) have computed the production rate of the oxidizing agents. From these computations two cases have been studied: a probable case with a production of 144 moles H_2O_2 in one million years and a hypothetical maximum case with a production of 29 000 moles H_2O_2 . In the first case the canister is assumed to be intersected by one fissure and in the other by five fissures.

The computations for the probable case give an extension of the redox front in the direction of flow 6.8 m in one million years, compared with 6 m obtained by the analytical solution. The extension in the transverse direction from the centre line has been calculated to be at most 1.6 m. The penetration into the rock matrix at the inlet of the fissure has been calculated to be $3.7 \cdot 10^{-2}$ m.

AN APPROACH TO MODELLING RADIONUCLIDE MIGRATION IN A MEDIUM WITH
STRONGLY VARYING VELOCITY AND BLOCK SIZES ALONG THE FLOW PATH

I Neretnieks, A Rasmuson
Royal Institute of Technology, May 1983

SUMMARY

Radionuclides escaping from a repository in crystalline rock for spent nuclear fuel will migrate with the seeping water in the fissures. Most radionuclides will be retarded by sorption on the rock surfaces and by diffusion into the rock matrix. Available surface for sorption and residence time are two prime variables which influence the radionuclide movement.

The water velocity may vary very much along a flow path especially if the flow path enters a strongly fissured zone (lineament).

In this paper the radionuclides in a stream tube with an arbitrary velocity along the flow path have been investigated and a numerical scheme based on the integrated finite difference method - IFDM - is proposed for practical calculations.

The transport mechanisms considered are advection and longitudinal dispersion and the retardation mechanisms used are instantaneous sorption in a portion of the rock (surface sorption) and diffusion into the rock matrix and sorption on the micropore surfaces.

An attempt is also made to account for blocks of various sizes by the MINC approach (Multiple Interacting Continua). This method accounts for the large sorption surface but small volume of small blocks and small surface area but large volume of large blocks. Any block size distribution can be handled.

ANALYSIS OF GROUNDWATER FROM DEEP BOREHOLES IN KAMLUNGE

Sif Laurent
Swedish Environmental Research Institute, May 1983

SUMMARY

Groundwater from four boreholes in granitic rock at an investigation site in Kamlunge has been sampled and analysed. This is part of a larger program of geological, geophysical and hydrogeological investigations aimed at finding a suitable site for a high level radioactive waste repository.

One to four water-bearing levels in each borehole between a depth of 100 and 600 m were selected. Prior to sampling, the water-bearing level is isolated between packer sleeves. The water is then pumped to the surface where sensitive parameters such as redox potential, pH, sulphide and oxygen content are measured electrochemically on the flowing water in a system isolated from the air. Water, filter and gas samples are sent to several laboratories for further analysis.

The present report is a presentation of the results of the groundwater analyses. The reliability of the results is discussed but there is no evaluation in relation to geology and hydrogeology. This report presents the basic results from the groundwater analyses to be further evaluated by experts in different fields.

GAS MIGRATION THROUGH BENTONITE CLAY

Roland Pusch, Thomas Forsberg
University of Luleå, May 31, 1983

SUMMARY

Hydrogen gas produced by irradiation of pore water in the highly compacted bentonite that surrounds the copper canisters according to the KBS 2 and 3 concepts, may escape from the clay/copper interface if the gas pressure is higher than the groundwater pressure. A reasonable physical model predicts that gas may penetrate wider "capillary" passages that actually exist in the very dense clay, although these passages are still of microscopic size. In the large majority of the clay voids, the capillary action is sufficient, however, to resist gas penetration. and this suggests that a possible mechanism of gas migration is that of a "finger-like" pattern of tortuous gas passages extending from the canisters if radiolysis takes place at all. Two series of experiments have been run at gas pressures up to about 10 MPa. Nitrogen as well as hydrogen were used in these tests which seem to confirm, in principle, the validity of the physical model.

ON THE THERMAL CONDUCTIVITY AND THERMAL DIFFUSIVITY OF HIGHLY
COMPACTED BENTONITE

Sven Knutsson
University of Luleå, October 1983

SUMMARY

The report concerns the thermal conductivity and the thermal diffusivity of highly compacted bentonite, which has been suggested as embedding substance in the Swedish concept for the disposition of highly radioactive unprocessed nuclear waste products.

The first part of the report describes the testing method used, which is called the Transient Hot Strip method (THS). By this, the thermal conductivity and the thermal diffusivity were determined simultaneously in a test which was performed within 10-15 seconds. The measuring unit was a small metal strip, which was placed in the center of the tested bentonite sample. Due to the short measuring time (10-15 sec), the risk of water redistribution due to thermal gradients was reduced to a minimum. This made the method well adapted for the determination of the thermal properties of the moist bentonite.

The experimental set-up, including the use of the swelling pressure oedometers in which the bentonite samples were mounted, with the metal strip in the center, are described.

The second part of the report, describes the experiments, which were performed on bentonite bodies, mostly cylindrical in shape with a diameter of 50 mm and a height of 20 mm.

From the tests of the air dry samples it was found that the thermal conductivity increased as the pressure on the sample increased. This was explained by an improved thermal contact between the measuring metal strip and the bentonite body at increasing pressure. The determined conductivities were compared with the values obtained from a theoretical model for the prediction of the thermal conductivity of moist geological materials. The agreement was good and the model was then used to calculate the thermal conductivity of the bentonite as water was taken up by the sample. The experimentally obtained conductivities during this process, agreed fairly well with the calculated values.

At low pressures, the thermal conductivity was found to be 0.83-1.08 W/m,K at a water content of 4.1% and a bulk density of 1.96-2.17 t/m³. At complete water saturation, the conductivity was estimated at 1.35-1.45 W/m,K at the bulk density of 2.0-2.1 t/m³, being representative for the ultimate conditions in a repository.

The mass heat capacity of the bentonite was found to be 0.96-1.05 J/g,K, which was slightly higher than the expected value that can be derived on values from the literature. The successfully increasing volumetric heat capacity during water uptake was also determined and this was predicted by the model used.

URANIUM, THORIUM AND RADIUM IN SOIL AND CROPS - CALCULATIONS OF
TRANSFER FACTORS

Sverker Evans, Studsvik Energiteknik AB
Åke Eriksson, Swedish University of Agricultural Sciences
June 1983

ABSTRACT

The distribution of the naturally occurring radionuclides uranium, thorium and radium in soil, plant material and drainage water was evaluated. The plant/soil concentration factors showed that very small fractions of the nuclides were available for the plants. The water/soil concentration factors were calculated; the nuclide content in drainage water generally indicated very low leaching rates. The distribution of the radionuclides was utilized with the aim to obtain reliable concentration factors which in turn could be used to calculate the transfer of nuclides within the agricultural ecosystem. Dose calculations were performed using plant/soil concentration factors based on geometric mean values.

FISSURE FILLINGS FROM GIDEÅ, CENTRAL SWEDEN

Eva-Lena Tullborg, Swedish Geological
Sven Åke Larson, Geological Survey of Sweden
August 1983

SUMMARY

This report presents results from a fissure filling mineral study from a KBS test site at Gideå, eastern Sweden. Most work has been concentrated on boreholes Gi 2 and Gi 4. The former borehole has been drilled within a discharge area and the latter within a recharge area.

The most frequent fissure filling minerals at Gideå are calcite, chlorite, pyrite, laumontite, stilbite, smectite, quartz and epidote. Potentially "young" minerals, i.e. minerals which can be formed during present conditions, are smectite, calcite and quartz. Equilibrium calculations show that these minerals can be stable in the water analysed from Gideå. In borehole Gi 2, the water exhibits a greater equilibrium with existing minerals in the fissures than water from Gi 4. This is due to the recharge of the water in Gi 4 and discharge of the water in Gi 2. When carbonate saturated water penetrated through the bedrock, calcite precipitated within the more superficial portions of the rock. However, both calcite and zeolite fissure fillings are associated with certain zones in the drill holes.

A large number of fractures can be concluded to be of post-Jotnian age. Stilbite, calcite and smectite are associated with these fractures. Laumontite can also be assumed to belong here. The dolerites in the area exhibit a high fracture frequency but a relatively low hydraulic conductivity. This is due to the fact that smectite is very prevalent in the dolerite fractures, sealing the fractures.

Several generations of fracture filling minerals (above all calcite) have been found in a single fracture. Both textural and chemical properties distinguish them, which makes it likely that there are large age differences between them.

Stable isotope analyses of fissure filling calcites indicate that there are three different groups of calcite. Most of the calcites could not have been precipitated from water of present-day isotope composition at Gideå, whereas recent calcite may be found in one of the groups. Another group of calcite was precipitated during hydrothermal conditions.

THE GEOCHEMICAL BEHAVIOR OF PROTACTINIUM 231 AND ITS CHOSEN GEOCHEMICAL
ANALOGUE THORIUM IN THE BIOSPHERE

Marianne Gillberg-Wickman
National Defence Research Institute, Umeå, Sweden March 1983

SUMMARY

To be able to judge whether protactinium 231 might represent a major contribution to the human radiation risk from high level radioactive waste a literature study of the geochemical behavior of protactinium has been made. The interest in protactinium determinations has, as far, been in the field of marine geochemistry and geochronology. These investigations show that thorium may be used as a chemical analogue.

The content of protactinium 231 is determined by the ^{235}U content and consequently the occurrence of protactinium in nature is directly associated to the geochemistry of uranium.

The pronounced hydrolytic tendency of protactinium and its great sorption and coprecipitation capacity ought to prevent or at least appreciably delay its transport from a back-filled nuclear waste vault to the uppermost surface of the earth. It also has a tendency to form colloids or particulates which may be strongly fixed on a rock surface. In adsorption and desorption processes kinetics must play an important role. Our knowledge in this field is quite limited.

Under the physico-chemical conditions in the sea, protactinium is rapidly scavenged from the water column by particulates. It accumulates in the sediments.

PROCEEDINGS OF THE THIRD SPENT FUEL WORKSHOP

Editor: Lars Werme
SKBF/KBS, Stockholm, Sweden March 1984

PREFACE

The multinational spent fuel workshops have been held annually since 1981. The purpose of these meetings has been to create an informal forum for discussions on experimental techniques for studying the corrosion/dissolution of spent fuel in aqueous media as well as the interpretation of such experiments.

The first and second workshops were held in Studsvik, Sweden, on the initiative of SKBF/KBS. The third workshop, held in Boston, Mass., was organized by Battelle PNL. In the workshops, the contributing organizations have been Battelle PNL, AECL/Whiteshell and SKBF/KBS and its consultants, mainly Studsvik. At the third workshop also a representative from CEA was present as an observer.

Up to now, no workshop proceedings have been issued. However, the spent fuel workshops apparently fill a need as an international forum for discussing questions concerning spent fuel behaviour in nuclear waste repositories. It has therefore been decided to issue proceedings from the meetings to give continuously the updated position of the research in this field.

In addition to establishing the position of the spent fuel studies, the workshop has also assumed the objective of recommending topics for further studies in this field.